

Reply to Reviewer Comment 2

RC2: 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 occurrence 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.

GH: I will mention in the introduction that the arrow notation is a widely used notation at least if the product-operator formalism is applied.

Moreover, as a consequence to the discussions concerning the indices in Eq. (22) and the related SI section 1.1, I will insert into 2.1 a paragraph dealing with the connection between the propagation rules (those with an arrow) and the superoperator-density operator equations like $\hat{\rho}(t) = \hat{U}\hat{\rho}_0$. This also serves to emphasise that the two forms of the representation of the time evolution take place in different spaces: The arrow notation uses an operator base, while the matrix notation uses a base of column matrices. This means that the corresponding matrices are transposed to each other because they originate from dual spaces. I hope that this will clear up the misunderstandings regarding the indexing in equation (22).

My access to the commutator equation systems comes from the requirement of having a Liouville-invariant subspace, i.e. a multiple application of the Liouvillian and therefore the commutator serves as a tool to ensure the L-invariance of the subspace. Then this criterion is used also to find such a subspace. I will emphasize this more in the subsections 3.1 and 3.2.

RC2: 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 \rightarrow is *maps to*.

GH: I agree that this section of the SI was inadequate to explain some relations from the Main Part. The purpose of this section is to prove that (i) the

Liouvillian matrix is the transposed coefficient matrix of the system of commutator equations, and (ii) the coefficient matrix of the propagation formulae is the transposed propagator matrix. I have rewritten this section completely in the form containing proposition and proof. I hope that this will enable the reader to see the purpose of the section and, most importantly, to see why some relations given in the Main Part exist. For the latter, I will insert some remarks at the relevant places in the Main Part.

RC2: It may also help to explicitly explain the different symbols used for abstract operators (such as $\hat{\rho}$) and their matrix representations (such as ρ).

GH: I will insert a paragraph explaining these different symbols.

Technical issues

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.
GH: Thank you, this has been corrected.
2. page 2, line 28: I suggest to replace *in this case* by *in this example*.
GH: This suggestion has been added.
3. page 2, line 31: Typo in *dipol-dipol*
GH: Corrected.
4. same line: I suggest to add *or* before *cross polarization*.
GH: This suggestion has been added.
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*.
GH: This suggestion has been added.
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.
GH: I know of no counterexample, but I know of no proof. So I replace the "cannot be" with "is often not".
7. page 3, line 54: *...an initial state $\rho_0 = \hat{I}_z$* . (Shouldn't the density operator carry a $\hat{}$?). 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 (\rightarrow). 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.

GH: I agree with that comment. At first I wanted to characterize a substitution with this arrow, but now I see the danger of confusion and will replace all arrows with equal signs. The missing hat has been added.

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 $-it$...*

GH: I have changed my inadequate formulation to this suggestion.

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

GH: Changed.

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

GH: Changed.

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

GH: Changed.

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.)

GH: Corrected.

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.*

GH: This suggestion has been added.

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

GH: Corrected.

15. page 5, line 122: The operator A^\wedge is missing its hat.

GH: Corrected.

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.

GH: Yes, that needs to be corrected.

17. page 7, line 172: extra *all*
GH: Corrected.
18. page 7, line 177: parenthesis not closed
GH: Corrected.
19. page 7, line 181: extra *above*
GH: Corrected.
20. page 7, lines 181–182: Shouldn't all N be replaced by n ?
GH: Yes, that needs to be corrected.
21. page 8, Eq. 22: Isn't the matrix U multiplied from the left, resulting in $\hat{A}_1 \cdot U_{11} + \hat{A}_2 \cdot U_{21} + \dots$ (inverted indices of U_{kl})?
GH: The coefficient matrix of the propagation formulae is the transposed propagator matrix, so the indices in Eq. (22) are correct. This relationship was not adequately mentioned in the original manuscript incl. SI. See the reply to the reviewer's comment on SI section 1.1 above. This SI section has been completely rewritten to emphasize this relationship.
22. page 9, line 219: I suggest *...appearing in Eq. (5) and (6)*.
GH: This suggestion has been added.
23. page 10, line 243: replace *estimate* by *calculate*
GH: This sentence has been changed due to insertion of a note that the Liouvillian is the transposed coefficient matrix of the commutator equations.
24. page 12, line 286: typo, it should probably read: *...with the amplitude*

$$\frac{D_{IS}^2}{\omega_{IS} + D_{IS}}$$

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

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

GH: Corrected.

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

GH: Corrected.

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

GH: Corrected.

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.

GH: N and n have the meaning that the reviewer assumed. I change the sentence to include an explanation for these variables. The $N/2$ in the exponent is set into parentheses.

30. page 22, line 461: What is $\omega_{1l,s}$?

GH: I have replaced this variable with mixed index by two separate terms ω_{1l} and ω_{1s} .

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

GH: This proposal was adopted.

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

GH: Corrected.

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

GH: This proposal was adopted.

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 ?*

GH: This proposal was adopted.

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

GH: Corrected.

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)

GH: The minus is added, the example number is changed to 2D-1.

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

GH: Corrected.

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 ...*

GH: This sentence was replaced.

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

GH: The in-line equation $q = \sqrt{a^2 + b^2}$ has been inserted in the line after Eq. (S14).

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

GH: Corrected.

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

GH: Corrected.

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

GH: Corrected.

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

GH: Yes; this has to be corrected.

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

GH: Corrected to "In other words"

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

GH: Corrected.

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

GH: Corrected.