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Dear Editor,

Please find below our reply to the comments of the reviewers.
We wish to thank all reviewers for their careful reading and their constructive comments on this manuscript.

Besides, I should mention that we have added funding details in the « Financial Support » section that must appear in the published version of the manuscript.

Sincerely,

Daniel Abergel



Response to Reviewer #5

Please find below our response to the comments. We thank the reviewer for careful reading of the manuscript.

It has been recently shown that the “thermalized” semi-classical approach used in conventional treatments of NMR relaxation, while reliable in the treatment of “standard” experiments become unreliable in systems, such as hyperpolarized states, that are far from equilibrium. Treatment of such open quantum-mechanical systems is generally carried out in the formalism of a Lindblad master equation. The authors Rodin and Abergel show that this is formally equivalent to the standard 2nd order perturbation approach within a fully quantum mechanical framework under the usual approximations used in the treatment of NMR relaxation. I feel that this work is important in marrying the conventional perturbation approach that most NMR spectroscopists are familiar with and the “more formal” approaches to open quantum systems. I feel that this manuscript serves as an important (and complete) pedagogical framework for students of NMR relaxation to appreciate both the mathematical tools and the concepts encoded therein. I congratulate the authors for having produced such a thorough analysis.

I have only a few (very) minor suggestions –

1. I assume that the title implies that the manuscript integrates all approaches to NMR relaxation “under the sun”, not sure that it is formally true though I agree that the authors marry two (major) seemingly orthogonal approaches.

Actually, this title was not meant to refer to some extensive review of the many formulations of relaxation theory. We just addressed some fundamental aspects of the quantum mechanical formulation of the problem to emphasize the fact that all these aspects have been identified, if not treated, in the early days of NMR. Thus, the question mark in the title is mostly a rhetorical one, as we fear that the answer is negative, although we are extremely pleased to see that it has aroused some interest.

2. Starting with (2), the equations are written in units of \hbar . Therefore, the appearance of \hbar in the Boltzmann terms e.g., in (36) makes things somewhat inconsistent.

The indication that $\hbar = 1$ has been added to the text.

3. Define L in (36) for completeness.

The definition of L as the trace of $\exp(\beta H_B)$ has been added to the manuscript.

4. For (11), the authors should use the appropriate direct product symbol.

Corrected

5. In (23) perhaps it is worth stating that the expansion is in the basis of irreducible tensor operators of rank q ; though not absolutely necessary, this is what is generally done for NMR.

We prefer to leave this point undefined, as it involves subtleties of notations that may obscure the demonstration and be misleading at some point.

6. There are several instances where it is said that a certain equation “writes” e.g., before (26). These occurrences should be replaced by “becomes”.

Done

7. After (20), I would replace “...these denote functions...” by “...these denote standard time-correlation functions rather than...”.

Done

8. After (45), I would replace the phrase “.. express a Kubo kind of relation..” by “...express a relation similar to those provided by Kubo ...”

9. Line 305, define trace as Tr as in other cases.

Done

10. The Goldman classic “Formal Theory” is from 2001 not 2021.

We warmly thank the reviewer for pointing out this unfortunate typo.
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