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Reply on RC1

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Author comment on "Mechanisms of coherent re-arrangement for long-lived spin order" by Florin Teleanu and Paul R. Vasos, Magn. Reson. Discuss., https://doi.org/10.5194/mr-2021-36-AC1, 2021

Thank you for reading the paper and for the kind observations. Our intention was to provide an example of NMR pulse sequence design rationale, taken from the history of LLS development. As we stated in the Letter to the Editor accompanying the Ms, as well as in the Introduction (line 41, page 2), the Ms does not aim to communicate new research results, thereby belonging, as the editors explain to us, in the 'review' area. The scope is to revisit contributions to LLS pulse sequences with a hindsight of 15 years *circa*.

Though we initially intended to revisit spin systems in the weak *J*-coupling regime and sequences that perform well in terms of translating longitudinal magnetisation to singlet in such systems, the reviewer's comments (and the fact that such systems guarantee the longest spin memory) prompted us to address these sequences that over-populate singlet states in challenging systems containing nuclei near magnetic equivalence (figure 3 of the revised Ms and paragraph starting with line 229). The construction of these sequences contains useful information for the reader. Density operators are used to describe conversions of coherences in the singlet-triplet operator basis, highlighting the dynamics of the singlet state population.

Following the reviewer's suggestion, we have added a numerical simulation using SpinDynamica to show the efficiency of singlet excitation using M2S, SLIC and $ZZ+ZQ_x$ pulse sequences in both the strongly- and weakly-coupled regimes. Moreover, we have added results concerning the relaxation rate constant of the operator, i.e., the time-dependence of the auto-relaxation rate constants of the density operator corresponding to LLS, but in the absence of radio-frequency fields, in the revised manuscript (lines 188-205 pages 7-8).

We have also analysed in the Ms the similarities and differences between LLCs and zero-quantum coherences, between which comparisons are often drawn, and discussed their distinct nature and properties.

Other comments:

- *An abstract should be a brief and clear summary of the article contents. The current abstract does not fit that description and contains many digressions and pieces of exposition which do not enlighten. It should be cut down by a large factor.*

- We addressed this issue and abridged the manuscript's abstract, as well as the introduction.

- *The article concerns itself exclusively with systems in the weakly coupled limit, which was the main focus of singlet-state research in the 2000's but has since been somewhat displaced by interest in strongly coupled and near-equivalent systems. I was rather disappointed that the article included so little discussion or review of methodology in that regime, which perhaps holds more current interest.*

- We have added (*vide supra*) a paragraph about pulse-sequences that were developed for the nearly-equivalent spin systems, which are the most promising in terms of magnetisation lifetime conservation. M2S and SLIC pulse sequences are discussed at lines 229-238 of the revised Ms as well as in Figure 3 and the corresponding caption; several other pulse sequences, fit for both the strong- and weak-coupling situations, are also mentioned.

- *page 4 includes the phrase "provided the two spins are rendered identical". I know what the authors mean, but this phrase is misleading. Two nuclei of the same isotopic species are, of course, always identical.*

- We have changed the expression to *"provided the chemical shift difference between the two spins is eclipsed by ample radio-frequency radiation or by cycling the main field"* in the paragraph starting at line 95 in the revised Ms.

- *page 6 includes the phrase "changing the chirality of the magnetic system". The issue of chirality and in general, the symmetries of molecules and associated fields, is a very complex and deep issue, and I am not sure the authors are able to underpin this rather casual statement by rigorous theory. If so, they should do so. If not, they should steer clear.*

- The Greek term was aimed to evoke the hands-like reciprocal orientation of the (I_zS_z,ZQ_x) vector pairs prior to and after ZQ_x rotation by 180 degrees (clarified in Figure 2 and caption in the revised Ms) triggered by the chemical-shift difference. When locked by radio-frequency, the two configurations have marked differences in relaxation behaviour.

- *the term "pulsation" is used in several places where "precession" is probably intended.*

- Indeed, we have corrected accordingly, thank you.