

Magn. Reson. Discuss., referee comment RC1
<https://doi.org/10.5194/mr-2022-17-RC1>, 2022
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Comment on mr-2022-17

Frédéric Mentink-Vigier (Referee)

Referee comment on "Intermolecular contributions, filtration effects and signal composition of SIFTER (single-frequency technique for refocusing)" by Agathe Vanas et al., Magn. Reson. Discuss., <https://doi.org/10.5194/mr-2022-17-RC1>, 2022

Intermolecular contributions, filtration effects and composition of the SIFTER signal

SIFTER is a pulse sequence that can be used to extract the dipolar coupling, and thus the distance, between two unpaired electrons. The pulse sequence uses a single frequency, unlike PELDOR. The article describes a model to explain the SIFTER experimental data to better understand how the presence of inter molecular dipolar couplings affects the evolution of the signal and its decay/background. The work is very interesting and thorough. In general, the article describes in detail the strategy to tackle the problem and I have not found major issues. However, I have found at times the text to be quite dense or unclear, and below are my suggestions to make the reading easier.

Section 2: When starting the description of SIFTER, it would be good to have an example of a SIFTER trace and how the pulses are changed. This would help the reader get into the pulse sequence without going back to previous articles. Could you also add the Hamiltonians you consider in each section

L105: "In the second parentheses the first term appears due to the time evolution of the first term in the first parentheses and vice versa." The sentence is a little convoluted. I suggest rephrasing it.

L140 "Such monoradical like signal" is not clear. While you discuss, monoradical before, the "like" is not clear. May be just say: Monoradical-like signal or invert the order of the paragraph describing what was observed first in Doll's work

L159: wl and wlm are confusing. My understanding is that there is some redundancy here though I do not think it matters. wl refers to the intramolecular dipolar coupling of the

biradical I but not the spins themselves, while I_m is for the coupling between two spins I and m belonging to two different molecules. I would use upper case W_{lm} for intramolecular, and lower case w_{lm} for intermolecular. This would ease understanding where intermolecular couplings contribute.

For Eq 9, Since you use the same approach as in 2.1, I would suggest, for consistency, to use $\sigma(2\tau_1 - \delta t)$ should be used not V .

L170 $\tan(\omega\tau_1) \cdot \cos(\omega\tau_1) \approx \omega\tau_1 \cdot \cos(\omega\tau_1)$ is a disturbing approximation. It is far worse than a simple Taylor expansion of \sin . I understand why you do this to have a product over all N , I would do it first, then apply the approximation of $\tan(x)$ as $\sim x$. Actually you explain this later, L258 "In equation (21) we can add the missing factor \cos "

Eq(14), it would be easy to have some numerical values for those terms using a random distribution of radicals and have the corresponding decay. Also in the weak coupling approximation the equation can be further simplified using a Taylor expansion of the \cos . Note: for simulations, a good random distribution could be made with `packmol`.

L195 and subsequent, these sentences could be represented on a figure to simplify the writing.

L215, a graphical representation would help as well.

Eq 15. If B corresponds to decay, then should not we see operators in this equation?

L 243 what do you mean by all the A-B coupling terms? This would explain the introduction of Q .

Eq 23, R is not an obvious choice for an operator, as it often represents relaxation.

L367, missing space. ".All"

Could you move the figures where they are first mentioned?