



Paris, October 7th 2025

Re: “Long-Lived States Involving a Manifold of Fluorine-19 Spins in Fluorinated Aliphatic Chains”, by Coline Wiame, Sebastiaan Van Dyck, Kirill Sheberstov, Aiky Razanahoera and Geoffrey Bodenhausen

Dear editor,

Please find our responses below.

Reviewer 1:

I consider it suitable for publication after some corrections / modifications:

- Even if a deeper study on CSA relaxation effects is the objective of the authors for another paper, I consider it disappointing for the CSA discussion (and request for data) to have been dropped completely from this paper. As someone interested in relaxation mechanisms, beyond learning from the paper that $TLLS < T1$ in this class of molecules, I really would like to have some more of a hint "why". Assuming the authors do have an extensive NMR department with access to a say a 300/400 MHz NMR, in addition to the 500 MHz spectrometer used in this work, a valuable addition to this story seems low-hanging fruit: a quick measurement of $TLLS/T1$ for one molecule at two fields would be enough.

We thank Michael for your persistence; we performed measurements at 300 MHz spectrometer. These results are now incorporated into the revised version of manuscript. Indeed, both $T1$ and $TLLS$ become slightly longer at the lower field, revealing a slight contribution from CSA.

- In Figure 5, the dataset for the binding-affinity study, I had to search for the meaning of "contrast" on the vertical axis of the plot. Eventually I found an equation in the main text (paragraph starting line 225) expressing " C_{LLS} " in terms of $R_{LLS}=1/T_{LLS}$ values with/without protein in solution. Please make this more explicit in the figure by doing the

following: (1) Label the Y axis as " C_1 , C_{LLS} ", (2) State directly that C is a normalized contrast that is bounded between 0 and 1, (or 0 and 100%), assuming R_{LLS} in the presence of the protein is faster than the baseline (are there any situations where negative contrast is expected?). (3) State the analogous equation for C_1 , in terms of T_1 .

Thank you, we followed your suggestions improving the figure, mentioning the normalization in the caption, and providing the equation for T_1 .

Sincerely,

Kirill Sheberstov