

Magn. Reson. Discuss., referee comment RC1  
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## Review Report

Anonymous Referee #1

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Referee comment on "Heteronuclear and homonuclear radio-frequency-driven recoupling" by Evgeny Nimerovsky et al., Magn. Reson. Discuss., <https://doi.org/10.5194/mr-2021-31-RC1>, 2021

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The paper describes the use of rotor-synchronized RFDR pulse trains on two nuclei for simultaneous heteronuclear and homonuclear polarization transfer. Such a sequence can be used to replace a heteronuclear polarization-transfer step (e.g., CP) and a consecutive homonuclear polarization transfer step by a single element that achieves both transfers. The new method is demonstrated using simulations and experimental data on SH3. At fast MAS or probably more likely for rf pulses that occupy a large part of the rotor cycle, heteronuclear and homonuclear transfer are almost identical while for slow MAS or rf pulses that are short compared to the rotor cycle, little transfer is observed.

In Figure 2, the CP and RFDR overlay spectrum is virtually invisible. Please improve the graphic representation. It would also be more interesting to see a 1D trace comparison of the HET-RFDR and the CP+RFDR spectrum and not two spectra at different mixing times. I think it would be nice to discuss the different transfer characteristics of the two sequences since there are marked differences in cross peak appearance.

Why is the transfer to the Ca in Fig. 3f negative? Is this a double-quantum type Hamiltonian that is generated here? Can you please comment in the paper whether the observed variations in the transfer efficiency are in agreement with expectations from the simulations/operator analysis?

Is there a reason why the rf-field amplitudes used in Figs. 4 and 5 are different? 83 vs 65 kHz? It would be much nicer to use the same rf field to make them directly comparable. I guess the simulation effort is not very high to change Fig. 5. I also think it would be nice to duplicate Fig. 5 in the SI for fast spinning so one can better understand the differences between the two cases in Fig. 4.