Magn. Reson. Discuss.,
https://doi.org/10.5194/mr-2020-18-RC2, 2020

Open Access MAGNETIC RESONANCE Discussions

Interactive comment on "Using nutation-frequency-selective pulses to reduce radio-frequency field inhomogeneity in solid-state NMR" *by* Kathrin Aebischer et al.

Paul Hodgkinson (Referee)

paul.hodgkinson@durham.ac.uk

Received and published: 5 August 2020

This is an excellent contribution to the difficult problem of understanding and optimising ^{1}H homonuclear decoupling in solid-state NMR.

The introduction gives an effective overview of prior work. I would add the method proposed by Odedra and Wimperis (DOI: 10.1016/j.jmr.2013.04.002) for quick-and-dirty imaging of the RF imhomogeneity using the z shim (i.e. not needing a magic-angle gradient). I feel it should be clearer that this work builds, conceptually at least, on the 2002 Charmont paper cited. This can be done by moving the sentence beginning "Alternatively" to the start of the final paragraph.

The theory is mostly clearly explained, although some wording is a bit unclear which meant I was reading some sentences a couple of times to understand the point being made. For example, we don't really "apply RF orthogonal to the static magnetic field" (at least not in MAS probes). The truncation of the Hamiltonian at high field could be more carefully expressed, particularly as this is important for the following step. I similarly didn't understand the term "Larmor-frequency axis" (a frequency can't have an axis). I wasn't entirely convinced by the analogy between the spin-lock frame and the (Larmor) rotating frame; it is clear that ω_1 is much smaller than the Larmor frequency, but much less obvious, e.g. Fig. 1(c), that ω_2 is much less than ω_1. The breakdown of this approximation is referred to, but it is not clear how this would manifest itself in practice. Wouldn't it show in Fig. 2 as deviations between the simulated profile with and without the spin-lock component?

In terms of the simulations, I didn't really understand the significance of the blue line. Is this trying to approximate the effects of RF imhomogeneity without assuming a particular RF profile?

The experiments are all clearly described. The disappearance of the modulation sidebands when using the selective excitation pulse (Fig. 5) is interesting. Could this simply be explained by the breaking the rotor cycle periodicity by the time-dependent (and unsynchronised) shaped pulse?

The results obtained from the two different probes (Figs 7 and 8) were strikingly different and I feel deserve more comment. In particular, the response of the zero-frequency artifacts is very different. The artifact is much larger in Fig. 7, but vanishes when the shaped pulse is used, whereas the artifact is much smaller in Fig. 8 and the changes are much more modest. Is the behaviour in Fig. 7 reproducible? It is not obvious why the selective excitation would strongly reduce zero-frequency artifacts, and given that this is highlighted in the conclusions, it would good to understand this point a little better. There is some repetition in this section, e.g. Hellwagner 2020 doesn't need to be cited twice to make the same point. (Similarly for Bloch-Seigert earlier.)

Figure 8 is quite hard to read. It is not clear why different modulation frequencies were used for different shaped pulse lengths.

I'm not convinced that the linewidth data in Tables 1 and 2 is that useful in the main text, since I doubt that the quantitative results will be very reproducible - certainly not to 1 Hz! For example, there is a large reduction in FWHM specifically for the 16.8 ppm peak in Fig. 7, while the peaks behave much more uniformly in Fig. 8. The width of the 16.8 ppm in Fig. 7 looks more like a phase artifact (perhaps associated with the homonuclear decoupling?) rather than a tail due to RF imhomogeneity. The interacting nature of effects is a major headache for these experiments, but this is why a more detailed discussion of the weight that can be put on the quantitative results would be helpful.

It's good that the data sets are available on request. It would be even better if they were available via an open data repository!

Minor typographical issues:

- Not all symbols are defined, e.g. is τ_p in Fig 3 the duration of the shaped pulse or the combined duration of spin-lock + shaped pulse prior to nutation measurement?
- Axes are conventionally italic, e.g. x.
- In line 18, there should probably be a comma after "NMR". As currently worded, the sentence implies that there are some MAS probes in which the coil is not close to the sample. Adding the comma creates a separate clarifying clause which applies to all small MAS probes (probably the intended meaning).

Paul Hodgkinson

Interactive comment on Magn. Reson. Discuss., https://doi.org/10.5194/mr-2020-18, 2020.