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Interactive comment on "Origin of the Residual Linewidth Under FSLG-Based Homonuclear Decoupling in MAS Solid-State NMR" *by* Johannes Hellwagner et al.

Johannes Hellwagner et al.

maer@ethz.ch

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One aspect that didn't come across clearly was which spin parameters were significant in the third order terms, particularly in section 4 where heteronuculear couplings are now involved. Various spin operators are quoted, but it is not always clear whether these cross terms are arising from homonuclear dipolar couplings or heteronuclear dipolar couplings, cross-terms with offsets (or even 1H CSA). The analytical section refers to multiple three-spin operators, but presumably not all the contributions to the 3rd order terms will involve 3 spins?

We have added a sentence at line 155 where the second-order three-spin terms are

discussed first to explain that these are commutator terms between two different dipolar couplings: " They all arise from commutators of one homonuclear dipolar coupling in the interaction frame of the effective field with another dipolar coupling where one spin is in common." The third-order terms are of course more complex and we only discuss the ones that appear in situations where no second-order terms are found, i.e. in the two-spin system (line 170). Since there is only a single homonuclear dipolar coupling, they are double-commutator terms of this interaction with itself. We have modified a sentence on line 200 to make this clearer: "It is obvious from Figure 2 that the third-order terms (double commutator terms of a single homonuclear dipolar coupling in the interaction frame of the effective field) do not vanish under FSLG-irradiation ..." We do not discuss at all cross terms with the CSA tensor since we believe that the dipolar couplings are the dominant interaction that determines the residual line width under homonuclear decoupling.

Hence, I found the discussion on P10 and P11 a bit confusing. What exactly is meant by "simulations with one and two dipolar couplings", a CH vs. a CH2 spin system or is it H3? Similarly, I had to read the caption of Fig 4 a couple of times, e.g. (A) and (B) are showing calculated splittings, rather than "simulations" or "Full scale linewidths". It would be better to say "without taking chemical-shift *scaling* into account" ("correction" is a bit vague and could refer to referencing).

When rereading this section, I have to agree that this was not very clearly formulated. We have modified the start of the discussion to make it clear that we talk about simulations in homonuclear three-spin systems where one or two dipolar couplings are non zero (line 242): " In order to validate the theoretical consideration of the pulse sequence, numerical simulations in a homonuclear three-spin systems were performed with both pulse implementations shown in Figure 3. To validate the contributions of second- and third-order terms discussed in the theory section, simulations using only one ($\delta_1 2 \neq 0$) and two non-zero dipolar couplings ($\delta_1 2 \neq 0, \delta_1 3 \neq 0$) in a three-spin system were performed." We also amended the figure caption as suggested and added the

homonuclear three-spin system to it to make it clearer.

Is Fig. 7 is showing 13C or 1H T2'? The sudden shift to heteronuclear systems is a little jarring. In this context, I must disagree with the characterisation in the introduction of our work on 3rd order contribution in heteronuclear systems (Tatton, 2012) as merely "qualitative"! There was greater emphasis on numerical simulation; analysis was confined to the simple LG, with the merit that simple expressions could be obtained for the 3rd order contributions, but it was certainly quantitative, and considered some of the same problems (influence of 13C heteronuclear couplings on 1H homonuclear decoupled spetra). So the two papers are nicely complementary; indeed the contribution of 60 Hz to the linewidth agrees well with the earlier work, not just Fig S2!

The measurements are proton T2' and we have added this to the caption and the main text. We agree that qualitative is not a good characterization of ref. (Tatton, 2012) and have deleted the qualifier. We have also added, as suggested by the referee, that the observed heteronuclear broadening agrees well not only with Fig. S2 but also with literature results and added the reference on line 325.

Figure 8: Is a pi pulse being used in experimental spectra to refocus shift evolution in the indirect dimension? Were the simulations of the full 2D experiment or a 1D version? It will not affect the conclusions, but you wouldn't experiment the two versions of the experiment to match up quantitatively.

The experimental spectra are traces along the indirect dimension of a two-dimensional spectrum. No pi pulse was used in these measurements since we wanted to see spectra with chemical shifts which would be refocused by a pi pulse. The simulations are 1D simulations. The main reason for measuring 2D spectra was that we wanted to avoid windowed FSLG detection that produces additional artifacts. In simulations this is not a concern since detection can be implemented instantaneously.

Minor detail: Methanol and Ethanol in Materials and Methods should be lower case.

Corrected.

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