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 1  Response to Reviewer 1:
 2
 3  Thank you for the positive comments and the detailed reading of the manuscript.
 4
 5  Q: Fig. 4 caption, 3rd line: deviations which become
 6
 7  A: corrected
 8
 9  Q: Fig. 6 explain the straight lines in the caption
10
11  A: The lines are a guide to the eye to show the linear correlation. They go through the
 …  point (0,0) and the data point for the lowest MAS frequency. We have added the following
 …  sentences to Figure caption 6: "The straight lines go through (0,0) and the line width at
 …  the slowest spinning frequency. They are meant as a guide to the eye for the linear
 …  correlation."
12
13  Q: Fig. 8 avoid repeat of bue and red in c to avoid confusion with colours used in b: how
 …  about green and orange instead?
14
15  A: We have changed the coloring scheme to avoid confusion.
16
17  Q: Can the SI use S1 etc labels for page numbers, sections, Tables and Figures.
18
19  A: We have changed the labelling of the SI to include the "S" for figures, pages and
 …  tables.
20
21  Q: SI Figure 2: line 4, space between set and between
22
23  A: corrected as well as another error in this caption.
24
25
26  Response to Reviewer 2:
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28  Q: ABMS, like proton-proton dipolar interactions, yield a MAS dependent line width.  How
 …  do the authors differentiate between the two effects.  The same applies to the MAS
 …  induced frequency shift (see e.g. Alla and Lippmaa, Chem. Phys. Lett. 1982, 87, 30-33;
 …  Samoson et al. Solid State Nucl. Magn. Res. 2001, 20, 130-136).  While reading through
 …  the paper, one gets the impression that dipole-dipole interactions dominate the proton
 …  line width.  The abstract / title should be modified into something like "Residual line
 …  width resulting from proton dipolar interactions in Magic-Angle Spinning Solid-State NMR"
 …  in order to avoid confusions.  To differentiate, some experimental data on the field
 …  dependence of the MAS dependent line width would be highly appreciated.
29
30  A: We agree that the title should include the fact that we exclusively discuss broadening
 …  from dipolar contributions. Therefore, we have changed the title to "Residual Dipolar
 …  Linewidth in Magic-Angle Spinning Proton Solid-State NMR". We have also added dipolar in
 …  the abstract in two locations. The influence of ABMS on line position and line width has
 …  been proven to be elusive in the literature. The way we understand this effect is that
 …  ABMS originates from the shape and finite dimensions of the crystallites present in the
 …  powder sample. MAS averages ABMS contributions from the isotropic susceptibility but not
 …  ABMS effects originating from the anisotropic part of the susceptibility. The reason for
 …  this is the fact that they can be described as the product of two second-rank tensors
 …  that generate rank-0-2-4 components. Therefore we expect a line shift due to anisotropic
 …  ABMS contributions, rank-2 that are averaged out and rank-4 that are scaled but not fully
 …  averaged out (see Alla and Lippmaa, Chem. Phys. Lett. 1982, 87, 30-33 section 3).
 …  However, the ABMS line width should scale with $P_4(\cos(\theta_m))$ and should, in
 …  principle, be MAS independent as should be the shift. They are both $B_0$ field dependent
 …  as is experimentally shown in Samoson et al. Solid State Nucl. Magn. Res. 2001, 20,
 …  130-136. If our understanding of the ABMS shifts is wrong, we would appreciate more
 …  pointers where we misunderstood the literature.
31  We have not included experimental data since we are mostly interested in the theoretical
 …  underlying mechanism of the changes in scaling of the MAS line width. We reference
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31… several papers that discuss this experimental finding in the introduction (around line
… 40): "Experimental observations of the residual homogeneous line width as a function of
… spinning frequency show that it can often be approximated by a linear correlation with
… the inverse of the spinning frequency with some deviation that indicate a partial inverse
… quadratic dependence (Nishiyama, 2016; Sternberg et al., 2018; Penzel et al., 2019;
… Schledorn et al., 2020). This has been attributed to third-order contributions to the
… effective Hamiltonian or to chemical-shift effects (Sternberg et al., 2018; Moutzouri et
… al., 2020)." We hope that this is sufficient.

32

33 Q: In the manuscript, the authors do a great job in discussing the MAS dependent 1H line
… width. Unfortunately, the contribution to the signal that is hidden in the Pake like
… pattern in the base line is not quantified. Can the authors give an estimate how the
… intensity is changed with MAS frequency using second-order Hamiltonian arguments ? This
… would be extremely interesting, since most solid-state NMR experiments are sensitivity-
… and not resolution-limited. The manuscript is very similar to a recent paper by Xue et
… al. (J. Phys. Chem. C 2018, 122, 16437). The authors should discuss theoretical versus
… computational approaches to yield an understanding of proton resonances in the
… solid-state. In the presented approach, the geometry is restricted to very few angles and
… distances. Is it possible to derive general laws if only a few spins are considered ?

34

35 A: The line width is easy to assess via an expansion of the moments but of course there
… are many distributions that produce the same (second) moment and an easy guess of the
… line height is not possible. Of course in the limit of many spins, we might get a
… Gaussian line and then we could predict the line height. However, to what extend this is
… true, we do not know yet and is part of our current research efforts.
36 The appendix (and also references cited in the paper) give analytical expressions for the
… second- and third-order Hamiltonian. Based on these expressions, one can calculate
… analytical solutions of the line width. However, the expressions become very complex
… functions of the distances and relative orientations of the couplings and it is not easy
… to get much insight from them beyond the form of the spin operators. This is the reason
… that we opted for a numerical implementation of the effective Hamiltonians. The strength
… of this method is that we can distinguish which order of the effective Hamiltonian
… contributes which is not accessible from a purely numerical simulation as in the work by
… Xue.

37

38 Q: While reading the paper, one gets the impression that a proton line can be infinitely
… narrow if only the MAS frequency is high enough. The authors should add an additional
… term to their equations which summarizes the contributions to line width that are not
… affected by MAS. At which MAS frequency does the 1/omega_r dependence break down ?

39

40 A: The homonuclear dipolar part of the line width that is discussed in the paper has no
… spinning-frequency independent term. Of course, other homogeneous parts of the line
… width, e.g., relaxation terms or chemical exchange and inhomogeneous terms, e.g., sample
… inhomogeneity or ABMS shifts might broaden the line in addition to the dipolar line width
… as discussed in the introduction (around line 25). It depends on the relative magnitude
… of the various contributions where the 1/omega_r dependence breaks down and no general
… rule can be given. Especially, sample inhomogeneity and ABMS contributions can vary over
… a large range of values.

41

42 Q: I am missing a paragraph in which the theoretical considerations are compared with
… experimental data, and correlate theory and experiment. The author did this for the MAS
… induced frequency shift shown in fig 8 which is very nice. However, I am missing a
… discussion on the line width. The data for phospho-serine exist (at least in the MAS
… range 70-160 kHz), and it should be straight forward to read out the line width after a
… line shape fit.

43

44 A: The isotropic line shift can be compared quite well because it looks like the dipolar
… contribution to the isotropic line shift is the dominating one. As discussed above, there
… are many contributions to the line broadening and we do not claim that we can predict the
… line width from one simple three-spin simulation. In addition, the focus of the paper is
… on understanding the spinning-frequency dependence of the line broadening and not
… predicting the line broadening. Therefore, we do not want to make comparison to

44… experimental data of the line width but give references to published data that illustrate
… the spinning frequency dependence.

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