1. Report of the First Referee

The authors compare nuclear spin characterization using ESEEM and DD sequences. Overall the work seems timely and appropriate for publication in PRB.

However, I think some sections and figures would benefit from further revisions. Below is a list of comments and inquiries:

1) In some places the authors refer to second order hyperfine coupling, I assume this is in regard to the Zeeman splitting of the central spin but this could be clarified in the text.

2) I am not sure whether this is intentional but the equations miss punctuation and equation references the brackets.

3) Some equations are hard to read because they overlap with the text of the next column.

4) Related to 1) additional details to derive the Hamiltonian in Eq. (1) would be helpful, for instance when does this equation hold, I believe $\Delta \gg$ hyperfine elements is assumed to neglect coupling to Sx and Sy and then the nuclear spins are rotated such that there occur no terms with Iy? Is there a difference in the derivation for S=1 and S=1/2? Maybe an appendix applying the secular approximation and the appropriate rotation to the frame leading to this form would be helpful for future readers.

5) Fig. 2 misses the labels for the subfigures the same applies to Fig. 3 where I think that additionally the heading of the top left panel should read "Electron Spin 1".

6) In Eq. (2) it would help to provide the state that is used for the calculation as well as an intuition why it is sufficient to include the exponential decay term after solving the dynamics (and when this is valid).

7) The last paragraph of App. B is very hard to read and would benefit from some work on the language.

8) Could the authors comment on whether they have an intuition whether adapted approaches would be feasible for systems with an additional central spin like the (15N)V center or defects with even larger spin like vanadium defects in silicon carbide?

9) Could the authors provide a short comment comparing to entanglement based approaches for nuclear spin control, e.g., https://www.nature.com/articles/ncomms14660?

Concluding, if the above points are addressed, I deem the article suited for publication within PRB.

2. Reply to the First Referee

We appreciate the comments made by the referee. We clarify each point as follows:

1. Second order hyperfine couplings means terms like $(\frac{A_{zz}}{\omega_L})^2$ and $(\frac{A_{zx}}{\omega_L})^2$. This is stated in the sentence: ((... the second-order term $\frac{A_{zx}^2}{\omega_L^2}$ can be neglected ...))

To clarify this point, we add the following explanation to the text: ((... which is the inner product of precession axes of a nuclear spin condition on the electron spin sublevel. This expression indicates the advantage of using DD sequence for sublevels where either s_0 or s_1 is zero, only first order parallel hyperfine coupling $\frac{A_{zz}}{\omega_L}$ play a role. On the other hand, in spin-1/2 systems ($s_0 = -\frac{1}{2}$ and $s_1 = \frac{1}{2}$), the first order hyperfine coupling vanishes and only second order parallel hyperfine coupling $(\frac{A_{zz}}{\omega_L})^2$ and second order perpendicular hyperfine coupling $(\frac{A_{zx}}{\omega_L})^2$ remain present. This indicates weak sensitivity of DD sequence on different nuclear spins...))

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3. Report of the Second Referee

The authors propose a blueprint for efficient nuclear spin characterization with color centers. The authors claim that they propose a more straightforward approach for determining the hyperfine interactions among each nuclear and the electron spin. The manuscript is written clearly by comparing dynamical decoupling (DD) and Electron Spin Echo Envelop Modulation (ESEEM), the numerical results are valid and may could be of use in other future experiments. I think the results obtained by the authors are of interest for Phys. Rev. B readers, so I support the publication in its present form.

As some minor remarks which the authors may take into account to beatify their study further, I would like to mention:

1) There are no a, b, c and d in Fig. 2, Fig. 3 and Fig.5. There is lack of definitions and units of "Hyperfine Parameters" in abscissa in Fig. 2 and Fig. 3. This sentence is confusing: "The first 23 hyperfine parameters are Azz and the second 23 parameters are Azx of the nuclear spin register." What are the specific meanings of hyperfine parameters along horizontal axis and vertical axis in Figs2?

2) Are there any reasons for the exponential decay part in gray for almost all the equations? It is a little bit strange, and the exponential decay part looks not in same size as well.

3) Styles of references [9], [14], [19] and [21] are not uniform with other references.

4. Reply to the Second Referee

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5. Report of the Third Referee

Zahedian et al. present a theoretical study on a the appropriateness of various methods of nuclear spin spectroscopy using electron spins in solid state materials as probes. They compare dynamical decoupling based techniques and consider how using a spin-1/2 system as a probe differs to the more common case of a spin-1 (such as the NV center in diamond). As the authors note, spin-1/2 electron spin systems are emerging in materials such as diamond and silicon and so expanding the existing nuclear spectroscopy machinery to this case will be of interest to the community.

The paper and its main result that correlation-style dynamical decoupling sequences are most appropriate for performing nuclear spin spectroscopy with spin-1/2 electron spin systems should be of interest to the community, and so in principle I think it is appropriate to publish in PRB. In parts I found the arguments a little unclear and so I would recommend some minor revisions before publication. Some specific points:

1) It is not clear to me why the correlation sequence performs much worse for the spin-1 probe (comparing Fig 2(c) and 3(c)) and there is not much discussion of this point in the text. This result also seems to conflict with Fig 5. It also does not seem like the spin-1 probe should be worse than spin-1/2 in this case. Also, what are the parameters used to generate the plots Fig 3c,d - is it simulating the 72 pulse DDESEEM?

2) It would be interesting to know, roughly, how the results of e.g. Fig 5 scale with properties such as probe T1 and T2. Does this strongly impact the optimal sequence choice? Some spin-1/2 systems may have short T1 times as they are able to cross relax with other g=2 spins present in the crystal (or on the diamond surface, for instance).

3) One goal of mapping e.g. a nuclear spin cluster is to also use the central electron spin to selectively manipulate individual nuclear spins to use as ancillae or implement quantum information protocols. Is it straightforward that the spectral resolution offered by the correlation style measurements also allow this sort of control? At minimum the extra time overhead from using the longer sequence would need to be considered in practice. Could the authors comment on the outlook here?

4) Minor comment on figures: some labels are missing and fig 4 appears a bit blurry.

6. Reply to the Third Referee

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