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Interactive comment

Interactive comment on "Room temperature hyperpolarization of polycrystalline samples with optically polarized triplet electrons: Pentacene or Nitrogen-Vacancy center in diamond?" *by* Koichiro Miyanishi et al.

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The manuscript by Miyanishi et al describes phenomenology, and detailed analysis, of 13C polarization enhancement by DNP in two systems: diamond and benzoic acid. The authors create athermal electron polarizations in electron triplet states via optical pumping, then drive that polarization to 13C nuclei via the "integrated solid effect," which in these cases means matching electron Rabi frequencies to nuclear Larmor frequencies. That the authors achieve modest polarizations on the 13C reservoir is not surprising; this manuscript is particularly noteworthy in that the both the electron and

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Discussion paper

nuclear resonances are interrogated and that the detailed mechanism is revealed by a detailed model. Readers that are interested in the "physics" of DNP in an accessible and thoughtful paper need look no further that this work.

For the authors consideration I include some comments below, though I am recommending the manuscript be published as is.

Lines 19-33: Often neglected in modern times is the extensive DNP work in "inorganic" semiconductors such as GaAs (conflict of interest: I am an author on some of these works). Deep conduction band photoexcited electrons, or photoexcited electrons captured at various defects, afford a surprising array of spin physics...I call attention to any of the papers by Meriles et. al. at CCNY.

Lines 54-56: Although later in the paper you press the advantage of photoexcited states for DNP, it might be worth emphasizing that point here: once the OP-DNP is accomplished, the effects of unpaired electron spins vanish and the full suite of high resolution solids NMR in diamagnetic materials becomes available.

Line 65: It is kind of the authors to call attention to the 2010 paper by King et al; though I note the mechanism by which nuclear hyperpolarization occurs in those high-filed pumping experiments has not been ascertained.

Section 2.1 is particularly well written and accessible.

Section 4.1 and Figure 3: These EPR results are particularly compelling in making the case for the ISE mechanism.

Lines 162-163: I believe the shuttling experiments from Ajoy et al may be the first optically polarized and 13C hyperpolarized measurements, albeit by shuttling the samples into a high field NMR system.

Line 199: It is interesting to compare the 13C diamond T1 values ($\sim$100 seconds) with those reported in https://doi.org/10.1038/s41467-019-13042-3. If I red the graphs correctly, the samples used in Ajoy et al have a T1 value of $\sim$50 seconds at that same field strength, consistent with the higher P1 spin density is the Ajoy samples.

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