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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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Dear Authors,

I very much enjoyed reading your work. In this context and it led to two questions regarding the resonance condition that you have employed.

First, in your work you appear to have chosen to achieve resonance for perfectly aligned electron spins. Misalignment then results in a loss of resonance and interferes with the polarisation process.

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Discussion paper

In Sec V of Q. Chen, I. Schwarz, F. Jelezko, A. Retzker and M.B. Plenio. Optical hyperpolarization of 13C nuclear spins in nanodiamond ensembles. Phys. Rev. B 92, 184420 (2015) we have also examined an alternative resonance condition chosen for NV orientations that are orthogonal on the external magentic field. Theory suggests that this choice leads to a higher robustness to misorientations as a larger fraction of NVs will be found within a certain range of rotation angles. This was tried out experimentally in J. Scheuer et al. Optically induced dynamic nuclear spin polarization in diamond. New J. Phys. 18, 013040 (2016) but no systematic comparison was done due to the limitation of that particular experiment.

It would be interesting to see whether your eperimental setup may allow for a more careful assessment of the merits of this choice of resonance condition.

Second, in the same work Q. Chen, I. Schwarz, F. Jelezko, A. Retzker and M.B. Plenio. Optical hyperpolarization of 13C nuclear spins in nanodiamond ensembles. Phys. Rev. B 92, 184420 (2015) we have compared a driving scheme in the |0> <-> |m=+1> transition with a scheme that uses a double quantum transition |m=-1> <-> |0> <-> |m=+1> which exhibits a significantly enhanced robustness to misalignments of the NV center. This benefits of this approach have not been tested yet experimentally and it would seem interesting to do so in your current experiment.

Yours sincerely Martin Plenio

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