Thank you reviewer 1. Below we provide a response to your comments. Our response is in red and the uploaded revised manuscript highlights the changes made.

Anonymous Referee #1

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This manuscript reports on the influence of the 14N solid effect on electron spectral diffusion profiles measured at nitroxide radical concentrations relevant for dynamic nuclear polarization. The authors approach the problem by combining considerations on spin dynamics with an empirical parametrized fitting model. This approach leads to an improvement compared to simulations disregarding the 14N solid effect and, indeed, at 20 mM concentration to reasonable agreement with experimental results. At an intermediate concentration of 10 mM, the model turns out to be simplistic. This is useful work, which improves understanding of electron spectral diffusion at high nitroxide radical concentrations. I recommend publication in Magnetic Resonance after minor revision that takes into account the following suggestions:

1. It is very awkward to report and use a nitroxide g tensor g = [2.0065, 2.0037, 1.9997], which is certainly wrong. The values reported earlier by Florent et al. were (almost) in line with expectations from numerous other reports and from quantum-chemical computations. The sentence quoting a "systematic error of 4 mT in the determination of the external magnetic field" is ambiguous, as it does not tell whether the error applies to Florent et al. or to the present work (it is the present work). You should be able to check this by using the proton Larmor frequency from ELDOR-detected NMR for calibration. It may not be necessary to repeat all computations (the anisotropy is correct, the absolute error too small to influence these simulations), but the issue should be reported in a clear way.

We agree with the reviewer, indeed the reported g values are off and this is because of an error in the determination of the magnetic field. Unfortunately the EDNMR spectra were recorded with a too low resolution (because of the wide spectral width covered) to track this. In retrospect, the field should have been calibrated with a high resolution EDNMR in the ^{1}H region when the measurements were done. The shift for the proton frequency is 0.17 MHz, which is very small compared to linewith observed. For the ^{14}N it is smaller and therefore small error in the nuclei Larmor frequencies are negligible. Similarly, as the anisotropy of **g** is correct this small shift does not affect the selected orientation and the other terms that depend on **g** because the energies and their differences depend on the product g_{eff}xB, where the error in B has been compensated in **g**. We now explain this in the manuscript in p. 7 as follows:

The *g*-values obtained from the EPR simulations and further used in the EDNMR simulations differ from those reported by Florent *et al* (Florent et al., 2011) (*g* = [2.00988, 2.00614, 2.00194]) as they compensate for an error of 4 mT in determination of B_0. These g-values where used to determine the selected orientations and to calculate g_{ef} in Eq. 2. Because the energies and their differences depend on the product $g_{eff}B_0$, where the error in B_0 has been compensated in **g**, they are not affected by the error in the field. The shift of 4 mT in B_0 for the proton frequency 0.17 MHz, which is very small compared to EDNMR linewidth. For ^{14}N it is smaller and therefore small error in the nuclei Larmor frequencies are negligible.

2. Please explain why TM can serve as a lower limit for T2mw (page 11, line 13, and

wouldn't T2ρ be a better choice?). This is not obvious to me.

You are correct, but we did not measure $T_2\rho$ and we deleted this part of the sentence to avoid confusion.

3. You claim (page 19, line 2) that the weaker 14N signals on the negative side of the allowed transition are presumably due to co-alignment of 14N and 1H hyperfine tensors. Did you test this?

This was a misplaced sentence and it is incorrect. We revised this part and wrote
Figure 4b shows the simulated 2D-ELDOR contour plot, which reproduces most of the features observed in the experimental contours. Some discrepancies can be observed in the intensities of the forbidden transition lines which can be attributed to the simplifications of the model.

4. You appear to cite every paper on W-band ELDOR-detected NMR on nitroxides (page 4, line 7/8), except for the very first one (DOI: 10.1016/S0009-2614(98)00765-9)

We apologize for this unintentional omission and added the reference in p. 4.

Typos:
Page 4, line 29: space missing between "0.6" and "mm"
Page 9, caption Figure 2: there is a surplus red "[** small m]"
Page 11, line 24: "signals reveal very weak signals" is awkward (signals are very weak)
Page 12, line 7: "three-spin calculation" (delete surplus "s")
Page 12, line 15: "we added reintroduced" should only read "we added"
Page 12, line 20: "on for the different forbidden transitions" shouldn't this read "one for each of the forbidden transitions"?

Thanks - we fixed all these.