

Magn. Reson. Discuss., author comment AC2
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Reply on RC2

Sarah R. Sweger et al.

Author comment on "The effect of spin polarization on double electron–electron resonance (DEER) spectroscopy" by Sarah R. Sweger et al., Magn. Reson. Discuss.,
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We appreciate the detailed summary and feedback of your review. In response to your last general comment, the results of the integration of Eq. 37 in (Marko, 2013) and S37 & S38 in the present paper disagree. Both papers make the same set of assumptions, however, (Marko, 2013) derives zero for the integration, whereas we found that, analytically and numerically, the result is actually non-zero. Our work therefore corrects an error in (Marko, 2013). Our responses concerning the specific comments are as follows:

1. A curve for the polarization with respect to temperature for W-band frequency has been added to Figure 1b.
2. This is likely a result of limitations in the range of the phase shifter for the G-band setup. To achieve the 90° change, in some cases, the probe had to be screwed in more tightly (slightly changing the path length to the resonator) leading to a slight change in the pump pulse efficiency and consequently to a slight change of the decay rate, $k = 0.046(3) \mu\text{s}^{-1}$ for 0° shift and $k = 0.053(3) \mu\text{s}^{-1}$ for 90° shift.
3. Line 126 has been modified to clarify the wording to indicate that for data at both temperatures, the field value was chosen based upon the maximum intensity point of a field sweep at the pump frequency.
4. A sentence has been added to the main text (Line 253) to address the relevance and extension of the theory to high-spin systems. Similarly, a line was added to S1 (below Eq. S29) indicating where in the theory would differ in samples that are not thermally polarized.

5. We would expect there to be effects from polarization on any of the mentioned experiments, however, the background theories are quite different from the work presented in this manuscript. Unfortunately, there is no way to comment on the extent of those effects without doing a detailed theoretical analysis which is beyond the scope of this work.

6. The model would hold in the case of inhomogeneity in k . As a test example we examine a fairly broad Gaussian distribution of k with mean of $1 \mu\text{s}^{-1}$ and a full width at half maximum of $0.6 \mu\text{s}^{-1}$, as shown in the top panel of the attached plot. The decay curves calculated by integrating over this k distribution and calculated for the central k value are nearly identical (bottom panel). This is valid for both the in-phase and the out-of-phase components. In particular, the amplitude of the out-of-phase component is barely affected. Therefore, we conclude that k inhomogeneity is likely not responsible for the observed mismatch between theory and experiment.

7. We have chosen not to move these sections to the main text. Our criterion for separating these sections is so that the focus of the main text is novel physical insight. Any mathematics that are known or not directly relevant to the physical insight were placed in the supplement. They are of course still accessible for those interested. We believe this organization of the content makes the main article accessible to a broad magnetic-resonance audience that may not not be deeply experienced with mathematical aspects of magnetic resonance.

Please also note the supplement to this comment:

<https://mr.copernicus.org/preprints/mr-2022-8/mr-2022-8-AC2-supplement.pdf>