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Interactive comment on "EPR Study of NO radicals encased in modified open C60 Fullerenes" *by* Klaus-Peter Dinse et al.

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We are grateful to the referee for the constructive and helpful comments on our manuscript. Below we respond point-by-point to the comments and include suggestions for corresponding revisions of the manuscript.

Referee 2
1) The abstract needs some re-writing, compared to the clarity of the paper itself the abstract seems confusing, at least to me.
Response: The abstract will be modified for clarification (see also Ref 1, 1)).

2) At the end of the introduction, the authors state that a smaller g3-value "deduced

by an analysis of a CW measurement, necessitated confirmation by pulse ESR experiments, better suited for the study of very broad spectra." I would challenge this view of the difference in CW EPR and pulse EPR data content. In the case described here, the g3 value of 0.2 (instead of 0.7 reported now in this paper) was deduced by fitting CW EPR spectra (if I understood correctly). In a way, ESE-detected EPR has a built in T2-filter that simplifies the spectra (reducing the broadness of the high-field region). I think the authors should point that out, as this is the real original point they have made here (or, if they think this is wrong, explain, why pulse EPR may be better suited).
Response: The referee is quite right in his remark that because of possible T_2 variations in the spectral range the true absorption line shape can be distorted in an FSE spectrum. However, we disagree with the referee about a "simplification", i.e. complete loss of the extreme high-field part, of the spectrum suggested to exist for g_3 = 0.225 in the FSE spectra. The T_2 determined by a 2-pulse echo sequence does not show any significant shortening towards the high-field end of the observed FSE spectrum, instead T_2 increases at temperature used for the FSE spectra with increasing field. Furthermore, the FSE spectrum shows a clear high-field shoulder which is however much too broad for being picked up by typically achievable B_0 modulation amplitudes. In addition we would like to point out that g_3 = 0.225 corresponds to B_0 beyond 3 T, while the cw spectra were recorded only up to 1.5 T. For clarification, a sentence will be added to the introduction at line 26 after ". . . very broad spectra":
"Although a T_2 variation as function of the external field can distort the shape of a pulse derived spectrum to some extent, difficulties in detecting extremely broad spectra with virtually absent changes within the typically achievable B_0 modulation amplitudes in cw ESR can lead to misinterpretations, in particular if the suggested spectrum extends a factor of two beyond the possible acquisition range."

3) Figure 1 clearly needs to be amended with a chemical structure of the cage molecules - the DFT structures should also be shown from a side view, not only the

top view.
Response 3): Figure 1 will be modified (see Referee 1, 3)).

4) Figure 4 (PEANUT): the authors should explain the three black lines in the graph.
Response: Figure and caption will be modified. The corresponding sentence in the caption will read:
"The dashed vertical lines indicate the expected nutation frequency distributions (Stoll et al., 1998) at the three principal *g* values for NO@C60-OH3 in Table 1 (X band)."

5) Last line on page 6:"level splitting" instead of "spitting": Thank you for spotting.

6) Figs. 5 and 6 should be combined in one figure as a) and b) or Fig 6 should be moved into the SI. Again Figs 7 and 8 should be combined into one figure as a) and b).
Response: Figures 5 and 6 as well as Figs. 7 and 8 will be combined in one figure, respectively. We also would combine Figs. 2 and 3 as well as Figs. A1 and A2 into one figure, respectively.

7) Page 10, beginning of 3.2: Would the authors expect a better resolution at intermediate field values when the frequencies for the ENDOR experiments are varied (through changes in g-strain?)?
Response: Best resolution is expected for van Hove orientations since in our case g matrix axes and hfi axes are expected to be collinear. This is true irrespective of the mw frequency used. For arbitrary orientations it is expected that at 34 GHz a better resolution in ENDOR would be obtained. The corresponding field range, however, was inaccessible for us due to low g values of interest here.

8) Figures 9, 10, 11: While I am usually all for original presentation of data, in this case I just think that plotting the three spectra in three rows above each other in Fig. 9 (without additional 3-D shift) would make it easier to see the "evolution" of orientation-dependent spectra. Then, Figs. 10 (and 11, I believe) could also be included in this figure.
Response: We prefer to keep the presentation of Fig. 9, however, will combine it with Fig. 10, but keep Fig. 11 separate.

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