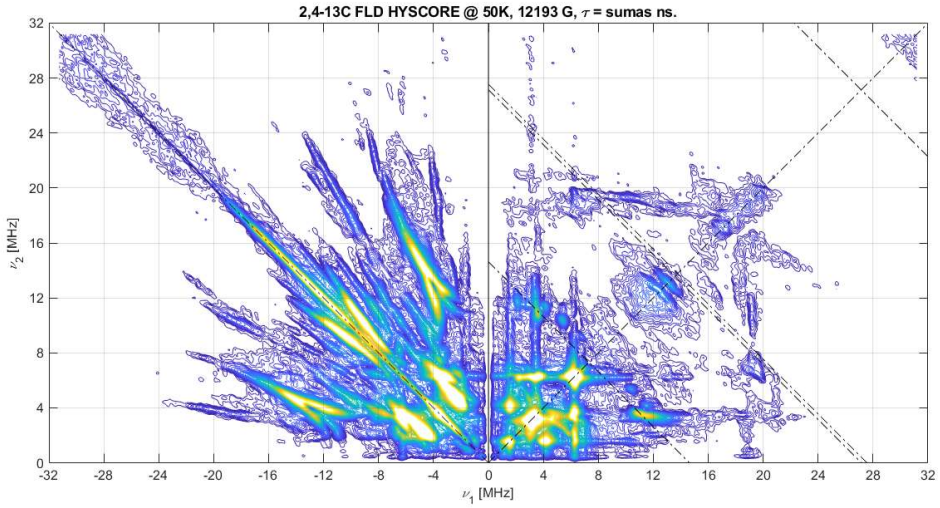
Reviewer 1

Section "Experimental parameters":

1.- Erratum in temperature (p. 362) has been corrected.

2.- HYSCORE experiments were measured at several tau values for the 13C-labelled samples (tau = 96, 112, 124, 176 and 144 ns for the central position, for example). In all of the spectra, the spectral features due to 13C(4) are very, very weak, yet, the spectrum shown in figure 4 of the manuscript (tau=112 ns) a bit better quality than the others. 13C features, which are very near the noise level show much better in the spectrum for tau=112 ns because it has a slightly better s/n ratio, that is why we decided to display it alone in the manuscript. The following figure, is the sum of the spectra corresponding to 124, 176 and 144 ns. As one can see, the 13C features are clearly there, but the spectrum in the manuscript is able to show them better. Moreover, no evident feature present in any other spectra is missing in the spectrum for tau 112 ns.



3.- To set and optimize the parameters, a single 1000 ns pulse was set and its power optimized to produce an FID. The FID integrated intensity was recorded as a function of the magnetic field. Then, the position of the magnetic field was fixed either at the center of the field-swept spectrum or at the high-end tail and an initial HTA ELDOR pulse was added. The ELDOR channel attenuation was initially set to 0 db, and several ELDOR-detected NMR spectra were taken varying the length of the ELDOR pulse from 1000 to 5000 ns. Then, the operation was repeated for several levels of ELDOR mw power. From the resulting spectra, the best s/n was found for HTA 1000 ns long and 0 db ELDOR power attenuation, so these parameters were adopted for longer accumulation of the spectra. The interpulse delay was chosen to be 1500 ns, long enough to let the potential FID of the first pulse decay. Information on the power of the HTA pulse and how the parameters were chosen has been added to the experimental section.

4.- Both, FID- detected and Echo-detected spectra were recorded. The referee is perfectly right, the FID-detected field-sweep experiments were recorded as a part of the set-up procedure, but in the figure, echo-detected experiments are shown, just to indicate the field position where the ELDOR-detected NMR were taken.

Section "Orientation Selección and Simulations:

1.- Yes, the calculations of the orientation selection patterns is based on the simulation of the Q-band CW spectrum. An X-band simulation is now shown in Fig. 2 together with the experimental spectra.

2.- Misleading comment on orientation selection in Fig. 3 caption and text has been changed. The orientation selection patterns calculated for Q-band frequencies, and shown in the figure show the orientations being excited for the spectra recorded at the maximum of the EPR absorption and the high-field tail. The patterns are, again, based on the simulation of the spectrum at Q-band frequencies, so, they should be reliable. This pattern, for the magnetic field set to the maximum of the spectrum shows that all orientations are excited. While the orientations in the plane of the flavin ring contribute more, there is still some contribution of the orientations close to the perpendicular to the plane.

3.- The position of the nuclear features on the HYSCORE spectra were first calculated taking into account single nuclei one by one using *endorfreq*. Then, the simulation of the spectra was done with *saffron* using the complete set of nuclei which want to be simulated in order to obtain the feature's intensities and combination lines. We have done some rephrasing in the manuscript in order to clarify this paragraph, the referee was right in pointing out some confusion.

4.- In the HYSCORE simulations using saffron, orientation selection was indeed considered.

Section "Experimental Hyperfina Spectra":

- Experimental features described in the text has been highlighted (Figs. 3 and 4).

Section "Error of measured parameters":

1.- Error in the expression (4) should be propagated from the individual errors to the subtraction. This is obtained from the squared root of the sum of the squared individual errors. The value $\pm$0.4 mT is properly calculated.

2.- We have added on page 18 a brief explanation on how the errors in the parameters of the Spin Hamiltonian were estimated. The errors for the estimated quadrupole couplings have been added as well.

Section "Discussion":

- Following the reviewer's suggestion, discussion has been shortened and restructured in order to make it clearer.

Section "Figure 1":

- Suggested changes in the position and information collected in Fig. 1 have been implemented.

Section "Spin Hamiltonian":

- SH equations and subindexes for tensor principal values have been corrected, simplified and explained.

Section “Further Comments”:

· Upon acceptance, raw data and simulation code will be made available in a open-access repository.

· Typos have been corrected.

· The paragraph describing results and analysis of EDNMR experiments has been rewritten in order to make it more clear (see also “ELDOR-detected NMR “suggestions of reviewer 2).

· The description of the EDNMR experiment (former 260-264 lines) was rephrased.

· It is true that the intensities of the 13C(4) simulations do not reproduce well the experimental spectra. However the position of the HYSCORE features already allows estimating the magnitude of the hyperfine coupling of the nucleus with high precision.