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Interactive comment on "Hyperfine spectroscopy in a quantum-limited spectrometer" *by* Sebastian Probst et al.

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The manuscript by Probst et al. reports the observation of electron-spin-echo-envelope modulations on their so-called quantum-limited spectrometer. As a follow-up to previous publications on this special high-sensitivity system, the authors demonstrate that they have realized the bandwidth required to detect the echo modulation owing to the interaction of an electron spin with very weakly coupled nuclear spins. This represents a next successful step in quantum-limited EPR and as such deserves publication.

As this referee report is part of a 'discussion' and follows the report by Graham Smith, I first note that I fully agree with his remarks/suggestions/questions. Taking these into consideration will for sure enhance the impact of the paper. This particularly applies to

the proposed discussion of the technical developments that are possible/ necessary/ foreseen. Doing so, the EPR community may be able to judge how close they (we) are to what the authors refer to as 'real-world applications'.

To clarify this point, I emphasize that ESEEM is a hyperfine spectroscopy. In other words, ESEEM is applied to probe the electronic wave function through the detection of the interaction of the electron spin with nuclear spins. For the two examples treated here, the erbium-doped CaWO4 and the bismuth donor in silicon, the observed modulations provide no information on the extent of the electronic wave function. For the erbium case, the erbium spin is (taken) completely localized on the erbium center, and the resolved ESEEM frequencies are determined by the positions of the tungsten nuclei, which are known from the crystal structure. For the bismuth case, modulations are at the Larmor frequency and refer to very weakly coupled silicon nuclei, so-called 'matrix nuclei', for which the ESEEM frequency does not deviate enough from the Larmor frequency to be resolved. In other words, these two examples suffice to demonstrate that ESEEM is feasible with this spectrometer, but do not generate any new information on the electronic wave function, i.e., do not fulfil the goal of hyperfine spectroscopy. These considerations are not meant to criticize the research, but to convince the authors that a discussion of the potential of the technique, in particular of the bandwidth that can be reached, may help to increase the impact of the paper.

Some minor points. 1. The authors might consider to shorten sections 2.1 to 2.3, because the description on page 2 to page 6 is standard and can be found in many textbooks. 2. For erbium, mI is described as good quantum number, which is not obvious for the experiments at the lower magnetic fields (cf. fig 3). 3. Sections numbered Arabic are referred to in the text by Roman numbers. 4. For the modeling of the data in fig. 7, the magnitude of the magnetic field is taken as a parameter. How do the resulting values compare with the experimental values? 5. To which experiment do the $\chi 2$ values in fig.9b refer? 6. Some numbers in figures may benefit from a larger font, e.g. in figures 4 and 11.