

#### Reviewer #1

We thank the reviewer for his positive evaluation of our manuscript. We have adjusted the figures following his comments.

For figure 1:

- font size has been adjusted,
- “hyperpolarization” stars have been removed to make the structure of the iridium complex more accessible,
- we have renamed the two hydrides (originally “H<sub>A</sub>” and “H<sub>X</sub>”) as “H<sub>I</sub>” and “H<sub>S</sub>”, for consistency with the spin operators used in figure 2,
- the structure of the co-substrate (mtz) has been added to the figure.

For figure 2:

- font size has been adjusted,
- pulses on H<sub>2</sub> are now explicitly displayed in (b),
- “J<sub>AX</sub>” has been renamed as “J<sub>IS</sub>” for consistency.

#### Reviewer #2

We appreciate the positive judgment about our manuscript. As suggested by the reviewer, the pulse sequence (Varian/Agilent) has been made available together with the raw and processed data as supplemental material. Further, a sketch of the experimental setup has been added as Appendix A.

#### Reviewer #3

We appreciate the positive evaluation of our manuscript. Following the advice of the reviewer:

- (1) we now explicitly report the magnetic field strength at which the NMR signal enhancement is referred,
- (2) we have added a 1D PHIP-NMR spectrum of the hydrides as Appendix B, indicating the frequency separation and the irradiation frequency,
- (3) having neglected cross-correlated relaxation in deriving equation 3-9 is now mentioned in the main text,
- (4) figure 2 has been modified to explicitly indicate selective pulses on H<sub>2</sub>.