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Reply on RC2

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Author comment on "Hyperpolarization and the physical boundary of Liouville space" by Malcolm H. Levitt and Christian Bengs, Magn. Reson. Discuss., https://doi.org/10.5194/mr-2021-26-AC3, 2021

I have a lot of sympathy with the referee's view. The term "polarization" does usually imply orientation of the magnetic moments in a particular external direction, so it is reasonable to restrict the term "hyperpolarization" to "an unusually large degree of orientation of the magnetic moments in a particular external direction", or something of that kind. Within this definition, "singlet (hyper)polarization" is self-contradictory, since the spins are not aligned with any external direction (although such a state may be interpreted as the spins in each pair having consistently opposite orientations). Tom Barbara, in the following comment, proposes the term "order" instead of "polarization", in which case one might introduce a new term "hyperordered" for states (such as strongly enhanced singlet order) which have a von Neumann entropy lower than that in thermal equilibrium, and which includes hyperpolarized states with greatly enhanced magnetization, but also low-entropy non-magnetic states such as that found in para-enriched hydrogen. I think that such a neologism would have a lot of merit.

Against that, however, I would balance the weight of current usage, in which "hyperpolarized" is often used in a sense broader than enhanced magnetization. That includes much of the NMR literature and also parts of atomic physics, where "hyperfine polarization" encompasses singlet order involving nuclei and unpaired electrons. Furthermore, the usage of words does shift in time, often to a place remote from their linguistic origins. For example, "nice" used to mean "stupid" and the term "orientation" originally meant "pointing towards the East", so one might raise an objection against spins being oriented with respect to the magnetic field of the earth, since that field never points East. In other words, etymology is not a good reference point for how words are used in practice. We have no Académie Française to lay down the law in such matters.

The referee says that the definition of hyperpolarization advocated in our article is "too broad to be useful" (at least I think that's what the referee means - it is not completely clear what the referee is referring to at that point). I disagree with the referee on that point. The von Neumann entropy criterion provides an unambiguous definition of hyperpolarization (or whatever other word is used), and we contend that is useful.

Personally I am open to introducing a term "hyperorder" as a generalization of "hyperpolarization" but I tend to think that it will not catch on and introduces a degree of unnecessary pedantry. Since Copernicus gives the opportunity for a community

discussion, I do wonder what other NMR scientists think about this.

To address the minor comments:

1. An ensemble of spins-I can support spherical tensor operators up to rank 2I. For example, spins-3/2 can have a maximum coherence order of 3, which may be represented as the m=3 component of a rank-3 spherical tensor operator. Together with the fixed value of the rank-0 polarization moment (eq.15), this gives a system of 2I+1 simultaneous equations, one for each of the spin states. We will clarify this in a manuscript revision.

2. I think the referee has misread Fig.4, The upper and lower scales on the figure refer to different quantities, related through eq.18b. The lower bound on the horizontal axis intersects at -1/(2sqrt3) =~ -0.288 (eq.32), which corresponds to pS= -1/3 through eq.21.

3. The cited article is very interesting and important but I do not immediately identify the relevance to eq.35.

4. This was a legacy from an old figure. Red has become grey.

5. Another semantic problem. I contend that the spin ensemble may have a "state" which is described by a density operator, just as a single spin system may have a "state" that is described by a Hilbert space ket. Nevertheless I agree it would be possible to replace "state" by "density operator" in this place.

6. The Lindbladian does also lead to thermal equilibrium, which is a point well off the left hand edge of the plotted region, corresponding to a zero projection onto the horizontal axis, and a projection of pzeq onto the vertical axis. That will be clarified in the text.