In their paper, Levitt and Bengs show that physical states characterizing nuclear spin systems are confined within a boundary of multidimensional simplexes in the Liouville space. The paper is very interesting and well written; there are only a few very minor comments (see below) with respect to the content. I recommend publication in OMR after minor revision.

The main issue, in my opinion, lies in the terminology rather than science and it is about the term “hyperpolarization”.

The authors start in the beginning by asking “is pure parahydrogen hyperpolarized?” and later replying “our answer is yes”. They later admit (page 17) that their “… definition of hyperpolarization makes no explicit mention of population difference, or the existence of a net magnetic moment in a certain direction.”

Let me be very clear. My concern is not about science, it is about terminology. I worry that the definition of hyperpolarization given by the authors goes against the semantics of the word “polarization” (as well as its origins, i.e., etymology).

Where does the term “polarization” come from? For a two-spin system, the situation is clear. Equilibrium polarization is a normalized population difference, \( P_{eq} = \frac{(p_a - p_b)}{(p_a + p_b)} \); where \( p_a \) and \( p_b \) are populations of levels for spin states “alpha” and “beta”, respectively and, in NMR, \( P_{eq} \) is a very small number, typically below \( 10^{-5} \). Strong polarization in NMR is usually termed “hyperpolarization”; the border of what % polarization is considered “hyper” is now beautifully connected to the von Neumann entropy in this paper. So, clearly, in a specific case of isolated spins 1/2, overpopulation of one level with respect to another creates oriented magnetization which can be measured. Both words “magnetization” and “polarization” usually imply a vector that has a specific orientation.

So, it totally makes sense for the spin-1/2 case to call deviation from zero population difference “polarization” because it means a specific orientation of some vector quantity (magnetization) in space. The term “polarization” beyond physics is often used to imply a “division into two sharply distinct opposites” (see https://dictionary.cambridge.org/dictionary/english/polarization, https://www.merriam-
webster.com/dictionary/polarization or https://www.thefreedictionary.com/polarization) which connects well with this idea. In almost all forms of common NMR, we measure magnetization or, more fundamentally, oscillating single-quantum coherences (i.e., one-spin-flip-at-a-time allowed transitions). Calling other overpopulated multipole moments “hyperpolarization” is not the best idea, especially ones with rank zero! The phrase “singlet polarization” is an oxymoron, in my opinion; it is an NMR jargon which, more likely, will make many young minds studying this subject very confused, at least at the begging of their “wondering in this region”. I note, once the definition of “polarization moments” is introduced (eq. 9), everything else follows from that. This is probably where the root of my dissatisfaction is.

There is a big asymmetry here, though. While I am in a position to criticize such a definition, I do not necessarily have a good alternative... If we go deep into semantics, then probably equating polarization with orientation (i.e., only rank 1 tensors in the multipole extension of the density matrix) would be a correct choice. Obviously, this would probably cause many disagreements, since a huge body of NMR literature already uses this term in a much broader sense. One should also note that hyperpolarization is usually defined with respect to the specific groups of spins (because in high-field NMR we can selectively measure signals from different types of spins), this corresponds to the enhanced magnetization corresponding to those spins.

If we accept for a moment that hyperpolarization refers only to NMR-observable states, other states that satisfy equations 38 and 39 and not magnetization can be called “latent” or “hidden” hyperpolarization. This would be useful in the context of parahydrogen experiments. Indeed, there, it makes sense to talk about the conversion of non-observable spin orders into observable spin orders. Thus, defining hyperpolarization only as enhanced magnetization (which can be converted from latent non-observable spin orders via a chemical reaction or by using special pulse sequences) would make sense. Otherwise, everything in PHIP/SABRE is hyperpolarization and the discussion is over: the term becomes too broad to be useful.

Minor comments:

- Could the author add more information on the condition in the eq. (23). Where exactly does it come from (i.e., why a sum of lambda from 1 to 2I)?

- In Figure 4, -1/3 is listed as a lower bound but one can clearly see that the intersect with the x-axis (representing a contribution of the rank 0 polarization moment) is lower than |-0.3| (while it should cross at |-0.33|). Is it a representation error?

- A similar analysis of the absence of information in the nuclear spin system was recently performed and could be mentioned with respect to the eq. (35): https://doi.org/10.1038/s41467-019-10787-9

- Page 17. “being inside the red region” – what exactly is the red region?

- In Figure 7, “hyperpolarized states” are mentioned. This is clearly NMR jargon. States can be overpopulated or depleted but not hyperpolarized.

- I am not sure I fully understand Figure 8. Why does not polarization come to the equilibrium polarization eventually, even in the case of the Lindbladian equation? From the graph, it looks like it cannot ever come there..