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Comment on mr-2022-9

Steffen Glaser (Referee)

Referee comment on "Visualization of dynamics in coupled multi-spin systems" by Jingyan Xu et al., Magn. Reson. Discuss., https://doi.org/10.5194/mr-2022-9-RC4, 2022

The stated aim of the manuscript is to extend the concept of angular momentum probability ("AMP") surfaces (which have been shown to be useful in the understanding of atomic physics experiments) by so-called angular momentum coherence ("AMC") surfaces. The authors show that this makes it possible to create three-dimensional graphical representations of the density operator of coupled spins, which is illustrated for three concrete NMR pulse sequences. The authors also show that the suggested visualization is complete in the sense that the density operator can be reconstructed from a full set of "AMP" and "AMC" surfaces. The mathematical basis of the visualization approach appears to be solid (but I agree with the comments of other reviewers that the presentation of the derivations and proofs should probably be adapted to the readers of Magnetic Resonance).

In my view, the most important weak point of the current manuscript is a thorough discussion of how the presented approach is related to similar visualization techniques that have been introduced before for the visualization of coupled spin/angular momentum dynamics. In particular, the readers (as well as the referees and the editor) will be interested to see what are truly novel aspects in terms of the visualization approach or novel applications and to give a proper account of closely related previous work. (Before I discuss these aspects in more detail below, I would like to point out that even if the visualization should be closely related (or even be essentially identical) to previously published approaches, I would still be in favor to publish a revised manuscript in which these points are considered, because as far as I see, the proposed visualization variant has not been applied to concrete NMR settings yet and it should be interesting and useful for the readers to see whether or not it could have advantages compared to other visualizations approaches.)

Point #1:

Before addressing the main point (the potential novelty of the "AMC" surface representation), I would like to briefly discuss the established "AMP" surface representation for uncoupled spins or atoms or molecules with arbitrary angular momentum (called F in the nomenclature used in the manuscript or J in other settings). I

think the proper context in which this work should be placed is the general field of phase space representations, at least by referring to the books by W. P. Schleich, *Quantum Optics in Phase Space* (Wiley, New York, 2001), C. K. Zachos, D. B. Fairlie, and T. L. Curtright, *Quantum Mechanics in Phase Space: An Overview with Selected Papers* (World Scientific, Singapore, 2005). F. E. Schroeck, Jr., *Quantum Mechanics on Phase Space* (Springer, New York, 2013). T. L. Curtright, D. B. Fairlie, and C. K. Zachos, *A Concise Treatise on Quantum Mechanics in Phase Space* (World Scientific, Singapore, 2014 and the recent review by R. P. Rundle and M. J. Everitt in Adv. Quantum Technol. 2100016 (2021). For a general discussion and comparison of different families of phase-space representations, I refer in particular to the recent paper (B. Koczor, R. Zeier, S. J. Glaser, Continuous Phase-Space Representations for Finite-Dimensional Quantum States and their Tomography", Phys. Rev. A 101, 022318, 2020) and references therein.

In (Koczor et al., 2020), the focus is the family of so-called s-parametrized phase-space functions of which the widely-used Glauber P function (with s=1), the Wigner W function (with s=0) and the Husimi Q function (with s=-1) are special cases. The paper gives an overview how the plethora of different finite- and infinite-dimensional phase space representations are related and can be mapped to each other. In particular, see Fig. 2 of (Koczor et al., 2020) for a graphical overview, section III on phase-space representations for spins and Ref. 76 (Stratonovich, 1956), Ref. 58 (Argawal, 1981), Ref. 77 (Varilly et al., 1989), and Ref. 57 (Brif et al, 1999).

As far as I can see (but please correct me if I am wrong), the definition of the "AMP" representation appears to be identical with the finite-dimensional version of the Husimi Q function, which to my knowledge was first defined (under different names) in the early 1980s (Argawal, 1981). Surprisingly, the close connection (if not identity) of the "AMP" surfaces and the Husimi Q function for finite-dimensional quantum systems seems to have gone unnoticed – or at least has apparently not been pointed out in the previous literature (and I need to apologize that before reviewing the current manuscript, I was not aware of the "AMP" representation and therefore we had not explicitly mentioned it in (Koczor et al., 2020).

So far, I have not received through the library the book (M. Auzinsh, D. Budker, S. Rochester, 2010) cited in the manuscript by Xu et al. and could not check if more details are given there on the relation between "AMP" surfaces and other phase-space representations. However, I found Simon Rochester's thesis from 2010 online, in which he points out that Chapters 2-5 of his thesis are largely identical to sections in the book, of which he is an author. In chapter 2 of his thesis (section 2.3.3), it is pointed out that the expansion of "AMP" functions in terms of spherical harmonics in Eq. 2.39 is "quite similar" to the Wigner function for angular momentum states given in Eq. 2.40 (from Dowling et al., 1994). I assume that this is also pointed out in the book (M. Auzinsh, D. Budker, S. Rochester, 2010). In this section of the thesis is also correctly stated that the essential difference between the expansion of the Wigner function and the "AMP" function is "that the contributions of polarization moments of various ranks are weighted differently", resulting in the fact that "AMP" functions are non-negative. However, this is exactly the property of the Husimi Q function, which has the same weighting factors as the "AMP" function and the same physical interpretation, see also (Koczor et al., 2020), where the s-dependent weighting factors are explicitly given for the family of s-parametrized phase-space functions (see Eq. 5 and Fig. 3).

To summarize point #1, I think it would be very beneficial to point out in the paper that the "AMP" functions are in fact identical with the Husimi Q function and to add corresponding original references and Ref. (Koczor et al., 2020), in order to avoid potential confusion that can arise if different names are used for the same concept in closely related scientific communities and to clearly define the relation between Husimi Q/"AMP" functions and other members of the family of s-parametrized phase-space functions. It would also be interesting to state in the manuscript whether or not my impression is true that the definition of (Argawal, 1981) predates the (identical) definition of what is called the "AMP" function.

Point #2:

This point concerns the question about the degree of novelty of the "AMC" surface representation. In the introduction, of the manuscript, the authors mention the "DROPS" representation introduced by (Garon et al., 2015), but state that "while the approach conveniently reflects the dynamics and symmetry of individual spins …, it is challenging to extract information from the drops representing systems of equivalent (or nearly equivalent) spins" and that the approach based on "AMP" and "AMC" surfaces is introduced "to address these limitations". This section raises several issues:

Point 2.1: To avoid any confusion of nomenclature, let me first address a minor point:

In the introduction of the manuscript, it is implied that "spin drops" is equivalent to the "DROPS" representation, which is not the case and the two concepts should be clearly distinguished:

"DROPS" is a general approach to visualize operators of general spin systems (coupled or not) that was introduced in the paper (Garon et al., 2015). In addition, the implementation of the "DROPS" representation and visualization (based both on the "LISA basis and the multipole basis") in a Mathematica package is publicly available by downloading the file "**DROPS_1.0.zip**" at https://www.ch.nat.tum.de/en/ocnmr/media-reports/downloads.

"SpinDrops" is the name of an interactive software package that is freely available for the community (see www.spindrops.org), which (in addition to other approaches) also provides the option to visualize the density operator (as well as Hamiltonians, propagators etc.) using a "DROPS" representation.

Point 2.2:

The “DROPS” representation is a general mapping between operators and a set of spherical functions (so-called droplets). As pointed out in (Garon et al., 2015), it is a Wigner-type (generalized) phase-space representations, which is applicable for arbitrary spin systems. In (Garon et al., 2015), the detailed mapping (which is based on symmetry-adapted spherical tensors) was explicitly presented for systems consisting of up to three coupled spins ½. More recently, explicit symmetry-adapted spherical tensors were constructed based on which systems consisting of up to six spins ½ as well as for coupled spins > ½, based on which arbitrary operators in such systems can be represented and visualized (see Leiner, Zeier, Glaser, "Symmetry-Adapted Decomposition of Tensor Operators and the Visualization of Coupled Spin Systems", J. Phys. A: Math. Theor. 53, 495301, 2020).

Both in (Garon et al., 2015) and in (Leiner et al, 2020), we focused on the “DROPS” representation based on the so-called “LISA” basis of spherical tensor operators (with defined **li**nearity, **s**ubsystem, and **a**uxiliary criteria, such as permutation symmetry), which is specifically constructed to visualize the individual spin contributions, which are relevant in most high-field NMR experiments. (In fact, due to the symmetry-adapted “LISA” basis, also magnetically equivalent spins can be efficiently represented).

However, in addition to the “LISA” basis, in section VII and VIII, appendix F, Tables II, VI and VII, Figures 6 and 12 of (Garon et al., 2015), we also explicitly and extensively discussed “DROPS” representations based on so-called multipole spherical tensor basis operators, generalizing a visualization introduced by Merkel et al. in 2008 (Ref. 18 in Garon et al., 2015). I did not have a chance to make an in-depth comparison yet, but I appears that this “DROPS” representation based on the multipole basis is essentially identical (or at least very closely related) to the “AMC” functions. Note that in particular the form of the “multipole operators” corresponding to transitions between blocks with different total angular momentum quantum numbers defined in Eq. (F1) in appendix F of (Garon et al., 2015) appears to be identical to the corresponding definition of the spherical tensor operators in Eq. (A1) in appendix A of the manuscript by Xu, Budker and Barskiy on which the definition of the “AMC” surface functions is based. In fact, both the “multipole operators defined by (Garon et al., 2015) and the spherical tensor operators in Eq. (A1) of the manuscript by Xu et al. differ from the tensor operators in the “LISA” basis in not having a defined particle number (i.e., “linearity”) and inducing a different grouping into droplets. One (apparently trivial) difference seems to be that the droplets corresponding to transitions from “F_l” to “F_k” and from “F_k” to “F_l” are separately displayed in droplets in of (Garon et al., 2018) and (Merkel et al., 2008), whereas they are merged in the manuscript by Xu et al, whereas droplets corresponding to zero-quantum phase phi=0 and phi=pi/2 are displayed separately. Another small difference is that (at least the droplets corresponding to the diagonal blocks in Fig. 1 are non-negative (corresponding to a Husimi Q function representing angular momentum pointing probabilities), whereas in the “DROPS” representation based on multipole operators, the droplets corresponding to the diagonal blocks can have negative values (corresponding to a Wigner function, representing the expectation values of so-called axial tensor operators). However, as discussed above in point #1 (and as indicated in the thesis of Simon Rochester and also discussed in (Koczor et al., 2020)), it is straight-forward to transform between a Husimi Q and a Wigner W representation by simply changing the rank-dependent weighting factors of the polarization moments.

To summarize point 2: It would be very helpful for the readers to clearly state how closely

the combined “AMP” and “AMC” surface representation is related to the “DROPS” representation based on multipole operators and to point out potential differences and whether or not the differences are significant. In particular, the following statements need to be corrected:

Line 30 “… it is challenging to extract information from the drops representing systems of equivalent (or nearly equivalent) spins.” This is clearly not the case, in particular if the DROPS representation based on multipole operators is applied.

Line 34: “… To address these limitations, we introduce …”. As pointed out above, the DROPS representation based on multipole operators does not have the stated limitations

For the presented NMR examples, comparison of the “AMP/AMC” with the standard DROPS representation based on the “LISA” basis and/or with the DROPS representation based on multipole operators is not mandatory, but could be quite useful (at least for one of the presented NMR examples), to make it possible for the readers to judge (potential) advantages or disadvantages of the different visualization approaches.