

Jury Member Report – Doctor of Philosophy thesis.

Name of Candidate: Grigory Starkov

PhD Program: Physics

Title of Thesis: SIMULATIONS OF HIGH TEMPERATURE SPIN DYNAMICS

Supervisor: Prof. Boris Fine

Co-advisor: Prof. Anatoly Dymarsky

Chair of PhD defense Jury: Professor Anton Andreev

Email:

Date of Thesis Defense: 16 October 2019

Name of the Reviewer: Prof. Alexei Buchachenko

X I confirm the absence of any conflict of interest

(Alternatively, Reviewer can formulate a possible conflict)

Signature:



Date: Sep 12, 2019

The purpose of this report is to obtain an independent review from the members of PhD defense Jury before the thesis defense. The members of PhD defense Jury are asked to submit signed copy of the report at least 30 days prior the thesis defense. The Reviewers are asked to bring a copy of the completed report to the thesis defense and to discuss the contents of each report with each other before the thesis defense.

If the reviewers have any queries about the thesis which they wish to raise in advance, please contact the Chair of the Jury.

Reviewer's Report

Reviewers report should contain the following items:

- Brief evaluation of the thesis quality and overall structure of the dissertation.
- The relevance of the topic of dissertation work to its actual content
- The relevance of the methods used in the dissertation
- The scientific significance of the results obtained and their compliance with the international level and current state of the art
- The relevance of the obtained results to applications (if applicable)
- The quality of publications

The summary of issues to be addressed before/during the thesis defense

The PhD Thesis presented by Grigory Starkov is devoted to the development of the hybrid quantum-classical method for simulating the short-time dynamics of the spin-1/2 lattices, or, more precisely, free induction decay (FID) curves and spectral lineshapes available from the NMR experiments. The basic idea of the method is to reduce the number of spins whose dynamics is considered quantumly by treating some classically. As the problem does not permit a standard way of the degree-of-freedom separation by their characteristic evolution timescales, the quantum and classical spin subsystems were segregated in a geometrical way. Special care were taken to retain the cross-correlation of two subsystems, which warrants the correct behavior of the method with respect to the size of quantum spin subsystem.

The structure of the Thesis perfectly matches its methodological goal. First chapter gives the problem statement in the context of the solid-state NMR method, defines basic equations and conditions and provides concise but clear overview of the available approaches to calculate or interpret FID dynamics. Since it covers some analytical and phenomenological approaches, important properties of the related dynamical correlations are simultaneously introduced. Properties of the classical and quantum ensembles at infinite temperature are exposed in the second chapter to introduce the notion of quantum typicality and Hilbert space sampling that makes the proposed method efficient. The method itself is formulated in chapter 3, which is followed by its careful assessment in the model 1D and 2D cases and for the 3D systems that model real crystalline materials (chapters 4 and 5, respectively). Chapter 6 adds on the analytical consideration of the hybrid correlation function to put this object on the same footing with its quantum and classical counterparts. Conclusions and outlook follow. Two appendices clarify the simulation protocols and explain the software developed.

I dare to say that the structure, content and presentation of the new method are excellent (though not ideal, see the comments below). Far from being trivial, the foundations of the methods are described clearly. Both mathematical and numerical ways of proving the accuracy of the method are presented in the convincing manner and compared, whenever possible, with the results of alternative approaches. Numerical examples considered ranges from the simple to rather complicated demonstrating the capability of the method and hinting some future perspectives. Clear recipes for internal uncertainty checks are presented. I especially like to acknowledge the material of chapter 6. It allows one to consider the hybrid method not only as an efficient numerical tool to overcome the quantum bottleneck of exponentially growing complexity, but

also as a source of physically meaningful results for analysis. Appendices make the work presented fully reproducible. Overall, the Thesis qualify Grigory Starkov's deep knowledge of underlying physics, strong skills in solving physical problems in both analytical and numerical ways, critical thinking of the results and clever approaches to writing.

Still, I have quite a few comments and questions on some research and presentation aspects.

1. The author introduces (p.2) the units with the Boltzmann constant $k_B = 1$. It looks a bit misleading, the more so $\beta = 1/k_B T$ is always used in what follows.
2. Equation (1.9) is one of the cornerstone of the approach, which allows one to assume energy conservation on the timescale of spin relaxation. On the other hand, some arguments relying on the long-time limit (e.g., p.50) are used. Is it possible that long-time behavior of the measured FID curves would be affected by energy relaxation?
3. *Chapter 2 essentially lacks the references. Is it well-known (for the happy few) or constitutes original results of the Thesis?*
4. *In chapter 5, the choice of basic parameters is not specified for "real-materials applications". Apparently, gyromagnetic ratios were set for bare nuclei and spin-spin coupling constants are derived from them within the truncated magnetic dipolar approximation. Is this choice appropriate as the effective magnetic parameters in the real materials may be different? Are the results sensitive to the parameter choice?*
5. Statistical uncertainty estimate appears on p.52 a bit unexpectedly. Perhaps it deserves some introduction and reference to Appendix A.3. By the way, it looks strange that the statistics of the Si and calcium fluorapatite simulation is not attested.
6. *On p.59, fig. 5.8 comes out of order. Better place it together with the fig. 5.4.*
7. *Last paragraph on p.65 was not clear to me and should be checked for consistency.*
8. Throughout the Thesis, prohibitive complexity of the rigorous quantum simulations is repeatedly mentioned. It would be instructive to give examples of current limitations from literature or author's own experience.

Reiterating on the perfectness of the Thesis as a qualification work in novel methodology, I feel some dissatisfaction with the lack of a bit broader view. Why it is so important to know NMR lineshapes with high degree of accuracy? Which new properties of materials one can uncover having such possibility? Can they reveal the possibility of dynamical control as a way towards quantum devices? Those obvious questions and related challenges remain unattended. Well, it is

up to the author to choose between broader or narrower discussion. The reason why I decided to comment on it is the Outlook section. In its present form, this section is out of the style of the whole work, where detailed exposure, careful analysis and convincing proofs are given for each significant statement. In the Outlook, only the prospects for diamond NV centers and neutron structure factors are somehow justified (though with mixing an object and an observable). The prospects given as bullet points and the last statement have no solid background neither as amenable for the proposed methodology nor as research challenges worthy of dealing with. *I strongly recommend to rewrite Outlook section stressing on the perspectives which can be concisely and clearly justified on a base of the material presented in the Thesis. Other prospects can be considered as challenges, but the essence and reason of a challenge should be given.* This basically brings the question of broader context back.

Despite the work is well and accurately written, quite a few misprints and formatting inaccuracies exist. E.g. Acknowledgments line 2, p.1 1.10, sec.1.2.3 1.6, p.23 1.2 (missing reference), p.34 1.4, p.38 1.7, p.43 1.25, etc.; behavior/behaviour, equation references with and without parentheses,... *Careful proofreading should be helpful to eliminate these and other imperfections.*

All the comments I have aim the presentation improvement. Overall positive impression of the work and of the skills of its author allows me to recommend the Thesis to public PhD defense after minor corrections.

Provisional Recommendation

I recommend that the candidate should defend the thesis by means of a formal thesis defense

*I recommend that the candidate should defend the thesis by means of a formal thesis defense only after appropriate changes would be introduced in candidate's thesis according to the recommendations of the present report (**highlighted above in italic for clarity**)*

The thesis is not acceptable and I recommend that the candidate be exempt from the formal thesis defense