

Review of manuscript submitted to JCP in Feb. 2018, the revised version was rejected in April 2018.

Reviewer #1 Evaluations:

Recommendation: Major revision

Reviewer #1 (Comments to the Author):

In this manuscript, Doster proposes a new mathematical model for neutron spectra of proteins. The model is of intriguing simplicity, and yields fits (Figs 3-5) of outstanding quality. There can be no doubt that such important work merits publication in JCP.

In the past, some of Doster's seminal contributions have been overlooked or undervalued because of their poor presentation. This is particularly true for his discovery of the central importance of methyl group rotation that was announced in passing in a method paper in *Physica B*.

With the present manuscript, this should not happen again. Therefore I urge the author to work hard to improve the text. And I ask the editor, if necessary, to allow **several iterations** until this manuscript is well organised, clear, and focussed.

How can the reviewer foresee "several iterations", this tells the authors that this referee will delay a publication of the manuscript. This manuscript is very well organized, but not in the "appropriate" way.

I find it perfectly legitimate to explain in any detail how the present data analysis and fit model are superior to currently discussed alternatives. However, this should not stand in the way of a self-contained, stringent exposition of the authors own, positive contribution. To make a specific suggestion: Shrink the introduction; only explain the basic facts and assumptions that are necessary to construct the model (4). **Remove all polemics**. In exchange, expand the discussion; add dedicated subsections that explain in how far the model (4) conflicts with energy landscapes, force constants, three or five classes of motion, Dianoux-Volino, etc. Of course, this is only a suggestion; *the author shall be free to reorganize his work in whatever way he chooses - but reorganize he should*.

Revised version:

My conviction that this work contains novel ideas and ultimately ought to appear must be very strong indeed - otherwise I would have refused to work through a manuscript in such a bad shape. See all the little and not so little points below. You authors have cost me a full working day.

What follows is a long polemic list of very minor formal points to support the bad shape:

The recommendation of referee 1 to "remove all polemics" has not been followed. The passage that culminates in "closely related" in quotation marks on p 12 is a scarcely veiled accusation of plagiarism. **Comparing two statements is not polemic.**

"Commercial preparations sometimes contain" on p 14 is innuendo. A sentence that assumes responsibility could read "We speculate that their commercial preparation contained".

I don't get the point. This statement is clear and not polemic, it does not improve by adding "speculate". In the preparation that was used by the cited authors by Sigma acetat was a major component of the buffer. I said this in my referee report. The author (Sokolov) suddenly claimed in the revised version to have removed acetat. Acetat has fast rotating methyl groups. Looking at the

data there is little room for "speculation". To correct errors or problems of the sample preparation is more important to the progress of Science than the reputation of an author.

P 1 announces "a generic dynamic model with two major components." The word "major" is out of place: Model (4) has no minor components. Distinguish the model from the physical processes it approximates. **Polemics!**

P 2, citation 3: not mentioned before, should be "a preceding paper". **OK**

P 2, citation 4-7: how can paper 4 from 1986 report on experiments performed in 1989?

paper 4 contains the sample preparation and characterization, used in the experiment in 1989, which is an essential part of understanding the neutron scattering results. It is obvious that the referee criticizes the "self citation" of the author, even if it is highly appropriate.

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P 2, after talking about "dynamic transitions", "the processes were assigned to ... state transitions ...": This is confusing. Which processes? Could one say "jumps" instead of "state transitions" to avoid confusion with "dynamic transitions"?

P 3, L 1, removing "from the inelastic spectrum" would make the sentence clearer. **Thank you**

P 3 L 3 vs L 4, 6: again, "transition" used for two completely different things. **ok**

P 3 "We employ" (blue): Incomprehensible mental leap. Not even a paragraph break would do, I fear.

Why cite refs 5,8 for a sentence that describes what is novel about the present work?

P 3 "Neutron diffraction ..." Why mention diffraction here? Why support basic facts about cross sections with research literature references? In this context, always specify whether "hydrogen" stands for D or H.

P 4 Provide original or/and textbook reference for "Placzek moment expansion".

P 4 "Gaussian approximation": **another opportunity for non-self citations**, e.g. Zorn.

P 4 **"Most bio-neutron": Cite reviews, not one of your own research papers.**

P 5 "type II" left over from version 1.

P 5 Mössbauer: why cite ref 3 in addition to the original work 18?

In his first review, self-citation was not mentioned, now it assumes a dominant role, although the citation list did not change. This idea was adopted from the second referee. His criticism is mostly about citations. Since 2001 I have been the victim of a powerful citation cartel, "Doster did not publish anything new since 1989" it was established by Zaccai, Frauenfelder, Smith et al., which is well documented in the literature. This explains the focus on citations without discussion the achievements of the manuscript. There is very little

comment on the main content of the paper. Finally the MD analyst questions the expertise of the experimentalists:

Debates in this field are skewed because each author only analyzes his own data set. When there are discrepancies, it is hard to know whether they come from the data, or from the analysis. If I were an editor, I would no longer accept any QENS paper unless the author discloses and deposits his full set of raw and refined data.

Comments of reviewer 2

Reviewer #2 Evaluations:

Recommendation: Reject

My main concern with the manuscript is that it does not present anything new. I did not find any new important results or interpretations, neither any new ideas, or solid overview of the field. It looks more as a compilation of earlier studies by the same author with significant ignorance of many other published results. Although the author is one of the pioneers in the use of neutron scattering spectroscopy to study protein dynamics, this field now has many groups with a lot of important publications. Yet, almost 40% of references are self-citations. So, it does not present a good overview of the field. Moreover, the presented analysis is also questionable (*why?, Fourier transform?*) and contradicts to what we know from NMR, MD-simulations and some other neutron scattering data analyses. *By contrast the results are compared with NMR data.*

As a conclusion, I still don't see any usefulness of this manuscript. I'm afraid it might be more misleading and provide disservice to the neutron scattering community by trying to simplify in a wrong way the description of complex protein dynamics. So, I cannot recommend the manuscript for a publication in any journal.

The referee did not understand the meaning and dramatic progress induced by this paper. Again he refers to self-citations, suggesting a membership to the anti-Doster citation cartel. The Editor follows his advice of Rejection although Reviewer 2 presented only unfounded general statements. Referee 1 suggested revision, but the Editor was not willing to involve a third reviewer, with experimental neutron scattering knowledge. This unfair behavior suggests hidden information exchange.

My conclusion is, that the title was too much against Energy landscape models of Hans Frauenfelder, which brakes a taboo. Second, we show, that methyl groups rotate even in a dry protein. In contrast, in simulations by M. Tarek it was concluded in 2003, using an in vacuo simulation of a protein, that methyl groups are arrested, in dry proteins. There could be a conflict of interests.