No case against scattering theory

Joachim Wuttke[a,1]

In a series of papers, Frauenfelder et al. (1–3) propose a radical reinterpretation of incoherent neutron scattering by complex systems, specifically by protein hydration water, drawing into doubt the "currently accepted model, used for >50 y" (3). Under this model they subsume not only assumptions about the scattering target (sample) but also the theory that connects sample and scattering signal. Effectively, they attack the insight (4) that the dynamic structure factor $S(q,\omega)$ (which they incorrectly call "the scattering intensity") abstracts from scattering kinematics and depends only on nuclear position operators acting on the sample. They claim that the established theory cannot account for the q dependence of elastic incoherent neutron scattering from isotropic samples. As a remedy, they suggest fits that account ad hoc for the recoil of the sample and the transit time of the neutron.

Frauenfelder et al. overlook that the isotropy of *n-p* scattering is broken by the setup of a scattering experiment: incoming beam direction and detector location define the scattering vector $\boldsymbol{q}$. Through $S(\boldsymbol{q},\omega)$, the measured cross-section depends on $\boldsymbol{q}$, or q for isotropic samples. The non-Gaussianity of $S(q,0)$ is long known (5) and can have various reasons (6). Faulty Gaussian extrapolations $S(q\to 0,0)<1$ do not disprove scattering theory.

Recoil is inherent in the accepted scattering theory [see ref. 7 for a particularly clear exposition] since $S(\boldsymbol{q},\omega)$ only involves transition probabilities that change the sample momentum by $-\hbar\boldsymbol{q}$. The recoil energy is not linear in $\boldsymbol{q}$, as posited in ref. 3, but quadratic. It is implicitly contained in $S(\boldsymbol{q},\omega)$, clearly appears in the limiting cases of an unbound scatterer or of large q (see, e.g., appendix G in ref. 8), and gives rise to a distinct factor when $S(\boldsymbol{q},\omega)$ is approximated for a classical system (9).

To account for transit times, Frauenfelder et al. request that neutrons be described as de Broglie wave packets (3). This is not explicitly done in standard texts because incident and scattered wave packet "share absolutely the same time dependence" (10). There are no cross terms: the scattering intensity, given as the squared modulus of a superposition of wave amplitudes turns out to be the sum of squared moduli of monochromatic wave amplitudes. Therefore, the established practice of accounting for finite energy resolution at the level of scattering intensities, not of wave functions, is perfectly adequate.

Thus, there is nothing wrong with the accepted scattering theory. The "woes" of ref. 3 are unfounded. The proposed heuristic injection of recoil and transit time only would make $S(q,\omega)$ inconsistent. With all this, nothing is said about the physical idea that quasielastic spectra might be explained by protein substate fluctuations. To make this idea verifiable, it needs to be cast into a mathematical model that yields $S(q,\omega)$ within the established correlation function formalism (4), without reversing scattering theory.

1 Frauenfelder H, Young RD, Fenimore PW (2013) Dynamics and the free-energy landscape of proteins, explored with the Mössbauer effect and quasi-elastic neutron scattering. *J Phys Chem B* 117:13301–13307.
2 Frauenfelder H, Fenimore PW, Young RD (2014) A wave-mechanical model of incoherent quasielastic scattering in complex systems. *Proc Natl Acad Sci USA* 111:12764–12768.
3 Frauenfelder H, Young RD, Fenimore PW (2017) The role of momentum transfer during incoherent neutron scattering is explained by the energy landscape model. *Proc Natl Acad Sci USA* 114:5130–5135.
4 van Hove L (1954) Correlations in space and time and Born approximation scattering in systems of interacting particles. *Phys Rev* 95:249–262.
5 Doster W, Cusack S, Petry W (1989) Dynamical transition of myoglobin revealed by inelastic neutron scattering. *Nature* 337:754–756.
6 Zorn R (2009) On the evaluation of neutron scattering elastic scan data. *Nucl Instrum Methods Phys Res A* 603:439–445.
7 Schober H (2014) An introduction to the theory of nuclear neutron scattering in condensed matter. *J Neutron Res* 17:109–357.
8 Furrer A, Mesot J, Strässle T (2009) *Neutron Scattering in Condensed Matter Physics* (World Scientific, Singapore).
9 Kneller GR (1994) Inelastic neutron scattering from classical systems—stationary phase approximation of the scattering law. *Mol Phys* 83:63–87.
10 Blügel S (2012) Scattering theory: Born series. *Scattering Methods for Condensed Matter Research: Towards Novel Applications at Future Sources*, eds Angst M, Brückel T, Richter D, Zorn R (Forschungszentrum Jülich, Jülich, Germany).

[a]Jülich Centre for Neutron Science at Heinz Maier-Leibnitz Zentrum, Forschungszentrum Jülich GmbH, 85747 Garching, Germany

Author contributions: J.W. designed research, performed research, and wrote the paper.
The author declares no conflict of interest.

[1]Email: j.wuttke@fz-juelich.de.

www.pnas.org/cgi/doi/10.1073/pnas.1710583114