REPLY TO DOSTER:

Franck–Condon and Van Hove formulation of quasielastic neutron scattering from complex systems

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Doster (1) criticizes a number of points in ref. 2, where a Franck–Condon-type spectroscopic formulation of incoherent neutron scattering is presented. My responses are given below.

First, the Franck–Condon formulation of incoherent neutron scattering does not contradict standard scattering theory, and it is even based on it. It merely starts from a nonstandard form of the intermediate scattering function, which is (probably) due to Wick (3), instead from the usual form that appears in the well-known paper by Van Hove (4). As long as the Van Hove form is considered in the quantum regime, both forms are completely equivalent. The two textbook models are presented to illustrate exactly this point. Wick's paper (3), which appeared in 1954, certainly belongs to the "classical references" in neutron scattering theory.

Second, the Franck–Condon picture is not limited to discrete vibrational energy spectra. A whole section in ref. 2 titled "QENS from Complex Systems" (QENS: quasielastic neutron scattering) is explicitly devoted to diffusive relaxation processes in molecular systems, assuming a continuous distribution of energy levels with a density $\rho(E)$.

Third, the fact that the energy transfer in typical QENS experiments fulfills $\hbar\omega \ll k_B T$ in the observed ω range and the dynamic structure factor is here therefore approximately symmetric, $S(q,\omega) \approx S(q,-\omega)$, does not imply that the scattering functions can be treated in the classical limit, $\hbar \to 0$, where $S^{(cl)}(q,\omega) = S^{(cl)}(q,-\omega)$ by definition (in typical powder samples or in solution the dynamic structure factor depends only on the absolute value q of the momentum transfer). Even if the scattering system can be described by the laws of classical mechanics, the above classical limit is only appropriate if recoil effects can be completely neglected, that is, if the recoil energy is negligible compared with the thermal energy, $\hbar^2|\mathbf{q}|^2/(2M) \ll k_B T$ (5, 6).

Fourth, the nonseparability of quasielastic and elastic scattering is an experimental problem and a consequence of the mathematical form of the intermediate scattering function for spatially constrained atomic motions. For systems with slow power-law relaxation, the dynamic structure factor at small energy transfers displays here a fusion of the elastic and quasielastic line. This fusion does, however, not imply that the elastic and quasielastic parts of the spectrum cannot be estimated by appropriate models and appropriate fitting procedures.

Finally, in ref. 2 a quantum-mechanical energy landscape picture is used, which is not the same as the classical kinetic energy landscape picture that is used in the articles by Frauenfelder et al. (7, 8). The quantum mechanical picture even supports Wuttke's comment (9) on these articles, in the sense that it is in complete agreement with standard quantum mechanical scattering theory.

It should also be noted that ref. 2 is not meant as a paper on proteins, although the concept of energy landscapes is used in the sense of energy spectra.

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7 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.
8 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.
9 Wuttke J (2017) No case against scattering theory. *Proc Natl Acad Sci USA* 114:E8318.