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Determination of the cooperativity length in glass forming liquids and polymers

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Although the idea of a ‘cooperativity’ length scale ξ related to the glass transition is now wide-spread, there is much less consensus on whether this length scale can be related to thermodynamic fluctuations and, if yes, whether one has to consider temperature fluctuations δT . The crucial experiment to this end has to compare values of ξ from ‘thermodynamic’ formulae to independent values from structural-dynamics experiments.

In a seminal paper, Ernst Donth proposed to assign a length scale to the AC-calorimetric relaxation time using the spatial resolution of quasielastic neutron scattering. The main problem is to find a range of relaxation times that is accessible by both methods. To achieve this, significant progress in the performance of NSE as well as AC calorimetry was required.

A first experiment of this kind was realised on a glass-forming liquid, propylene glycol (PG). The result was that agreement with the thermodynamic calculations was better if temperature fluctuations were accounted for. Nevertheless, in PG the difference between the two thermodynamic estimates is small. Therefore, a material with a higher ‘contrast’ between the alternatives was used in a second experiment, poly(ethylmethacrylate) (PEMA). In addition, this experiment benefitted from the upgrade of J-NSE with superconducting coils. The new results show a clear agreement with the thermodynamic calculation involving temperature fluctuations.

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