372c Towards More Realistic Nonequilibrium Molecular Dynamics Simulations

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Nonequilibrium molecular dynamics (NEMD) simulations are a very valuable tool since they allow to directly observe the response of a system subjected to an external field.

These simulations generally take places according to the three following steps: (1) apply the external field (shear rate, electric field...), (2) wait for the system to reach a steady-state properties and (3) average the properties of interests in the steady state.

Step 2 implies that some thermostatting mechanism is used to account for heat dissipation and to allow the system to relax towards a steady state. Thermostats currently used result in the systematic and spurious onset of ordered phases for strong external fields [1]. We provide an alternative approach, based on a configurational expression for the temperature, which solves that problem.

Step 3 only gives satisfactory results if the external field is very strong (typically several orders of magnitude larger than those used in experiments on molecular fluids). Using the transient-time correlation function formalism, we show how NEMD simulations can be extended to realistic shear rates and electric fields.

[1] J. Delhommelle, Phys. Rev. E 71, 016705 (2005)