Exploring the origin of nonlinearity in microrheologically driven glass-formers via potential energy landscape analysis

Carsten F. E. Schroer* and Andreas Heuer†
Westfälische Wilhelms-Universität Münster and
NRW Graduate School of Chemistry
(Dated: September 13, 2013)

We perform computer simulations of a binary mixture of Lennard-Jones particles in which one particle is continuously pulled by a constant external force. For this stationary non-equilibrium system one observes a linear dependence of drift velocity on the applied force for low force strengths but a strongly nonlinear one for increasing forces. Interestingly, the nonlinearity of the dynamical response becomes more pronounced as the system approaches the glass-transition temperature.

In our former work[a], we introduced the concept of relating the dynamics of driven glass-forming systems to escape processes out of mesoscopic regions of the underlying potential energy landscape, so called *metabasins*. This analysis allows one to understand the dynamics of supercooled liquids as a continuous time random walk (CTRW) in configuration space, which is characterized by a distribution of waiting-times between two processes and spatial displacements. Surprisingly it turns out, that the nonlinear effects are mainly included in the temporal part of the CTRW while the spatial part basically obeys the linear response prediction.

Our recent goal is to go beyond the CTRW picture to understand, which properties of the underlying potential energy landscape are changing with the onset of nonlinear dynamics. In this context we can observe that both, the Boltzmann distribution of energies of the driven system as well as the local escape rates out of single metabasins change due to the applied force. While the first effect is closely connected to the studies of rejuvenation of previously aged glasses under shear, the latter one can be quantitatively understood by using the ansatz of a modified periodic cosine potential.

* c.schroer@uni-muenster.de
† andheuer@uni-muenster.de

[a] CFES and A. Heuer, J. Chem. Phys. **138**, 12A518 (2013)