Controlling the structure, phase behavior and dynamics of soft colloids by active interaction switching

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We explore the microstructure and phase behavior of confined soft colloids which can actively switch their interactions at a predefined kinetic rate. For this, we employ a Reactive Dynamical Density Functional Theory (R-DDFT) and reactive Brownian-Dynamics computer simulations (R-BD), and study the effect of a two-state switching of the size of colloids interacting with a Gaussian pair potential in the nonequilibrium steady state. The switching rate interpolates between a near-equilibrium binary mixture at low rates and a nonequilibrium monodisperse liquid for large rates, strongly affecting the one-body density profiles, adsorption, and pressure at confining walls. Importantly, we show that sufficiently fast switching impedes the phase separation of an (in equilibrium) unstable liquid, allowing the control of the degree of mixing and condensation and local microstructuring in a cellular confinement by tuning the switching rate [1, 2].

Our results also demonstrate that switching activity significantly modifies the non-equilibrium dynamics and diffusion coefficients of the individual particles, leading to a crossover from short to long times, with a regime for intermediate times showing anomalous diffusion [3]. In addition, the self-part of the van Hove function has a Gaussian form at short and long times, but becomes non-Gaussian at intermediates ones, having a crossover between short and large displacements. The corresponding self-intermediate scattering function shows the two-step relaxation patters typically observed in soft materials with heterogeneous dynamics such as glasses and gels. We also introduce a phenomenological Continuous Time Random Walk (CTRW) theory to understand the heterogeneous diffusion of this system. R-DDFT results are in excellent agreement with R-BD simulations and the analytical predictions of CTRW theory, thus confirming that R-DDFT constitutes a powerful method to investigate not only the structure and phase behavior, but also the dynamical properties of non-equilibrium active switching colloidal suspensions.

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[1] A. Moncho-Jordá and J. Dzubiella, *Controlling the Microstructure and Phase Behavior of Confined Soft Colloids by Active Interaction Switching*, Phys. Rev. Lett **125**, 078001 (2020).

[2] M. Bley, J. Dzubiella, and A. Moncho-Jordá, *Active binary switching of soft colloids: stability and structural properties*, Soft Matter **17**, 7682 (2021).

[3] M. Bley, P.I. Hurtado, J. Dzubiella, and A. Moncho-Jordá *Active interaction switching controls the dynamic heterogeneity of soft colloidal dispersions*, Soft Matter **18**, 397 (2022).