Microrheology of isotropic and anisotropic colloidal suspensions via dynamic Monte Carlo simulations

Fabin A. Garca Daza[1], Antonio M. Puertas[2], Alejandro Cuetos[3], and Alessandro Patti[1,4]
[1]Department of Chemical Engineering, The University of Manchester, Manchester, M13 9PL, UK
[2]Department of Chemistry and Physics, University of Almería, 04120, Almería, Spain
[3]Department of Physical, Chemical and Natural Systems, Pablo de Olavide University, 41013, Sevilla, Spain
[4]Department of Applied Physics, University of Granada, Campus Fuentenueva s/n, 18071 Granada, Spain

The study of the rheological properties of colloidal suspensions is of key relevance in a variety of systems involved in technological applications, including polymers, micelles, emulsions, and electrorheological fluids, among others. Over the last years, microrheology (MR) has emerged as a new methodology to investigate the rheological properties of these soft materials at the particle scale [1]. In particular, MR allows one to link the bulk rheology of fluids to the microscopic dynamics of probe particles (tracers) that freely diffuse in the host medium (passive MR) or are subjected to external forces (active MR).

In this work, we have applied and modified our dynamic Monte Carlo (DMC) simulation technique to investigate passive and active MR in isotropic and liquid-crystalline colloidal suspensions. As a preliminary step, the original DMC methodology [2, 3] has been adapted to investigate the Brownian motion of a tracer particle in a bath of colloidal particles (passive MR) and to describe the impact of an external force pulling a tracer across a colloidal suspension (active MR) [4]. While in passive MR the elastic (G') and viscous (G'') moduli of the host phase are derived from the tracer's mean squared displacement, in active MR the response of the tracer to the external force provides the possibility of exploring the nonlinear viscoelastic properties of the material. We simulate the dynamics of a spherical tracer in a bath of rod-like particles in isotropic, nematic and smectic phases to explore, from the point of view of passive MR, the effect of order of the host phase and the size of the tracer on microviscosity and on G' and G''. According to our results, the computation of the loss factor (G''/G') suggests that dissipation dominates over elasticity for both isotropic and liquid-crystalline phases. Additionally, we observe a progressive increase in the effective viscosity experienced by the tracer which depends on the tracer's size. Nevertheless, the results also suggest that significantly increasing the size of the tracer reduces its ability to recognise the local structure of the surrounding medium thus perceiving it as a continuous fluid with no apparent short- or long-range order. We also investigate through active MR the linear and non-linear response regimes in a bath of spheres and rod-like particles at different densities by evaluating the effect of a constant force acting on the tracer particle. The calculated effective friction coefficients exhibit a plateau at small and large values of the external force, while a non-linear regime is observed at intermediate forces which is caused by the appearance of force-thinning. Our DMC results are benchmarked against earlier Langevin dynamics simulations [1, 5] and, where possible, also against theoretical models [6].

[1] A. M. Puertas and T. Voigtmann, *Microrheology of colloidal systems*, J. Phys. Condens. Matter, **26**, 243101 (2014).

[2] A. Patti and A. Cuetos, *Brownian dynamics and dynamic Monte Carlo simulations of isotropic and liquid crystal phases of anisotropic colloidal particles: A comparative study*, Phys. Rev. E, **86**, 011403 (2012).

[3] A. Cuetos and A. Patti, *Equivalence of Brownian dynamics and dynamic Monte Carlo simulations in multicomponent colloidal suspensions*, Phys. Rev. E, **92**, 022302 (2015).

[4] F. A. Garca Daza, A. M. Puertas, A. Cuetos and A. Patti, *Microrheology of colloidal suspensions via dynamic Monte Carlo simulations*, J. Colloid Interface Sci. **605**, 182 (2022).

[5] I. Gazuz et al., *Active and Nonlinear Microrheology in dense colloidal suspensions*, Phys. Rev. Lett., **102**, 248302 (2009).

[6] T. M. Squires and J. F. Brady, *A simple paradigm for active and nonlinear microrheology*, Phys. Fluids, **17**, 073101 (2005).