

Hydrodynamic fluctuations in quasi-two dimensional diffusion

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We study the diffusion of colloids on a fluid-fluid interface using particle simulations and fluctuating hydrodynamics. Diffusion of colloidal particles confined to a surface is a key transport mechanism in many technological and biological systems. For example, it is known that the transverse diffusion of proteins embedded in lipid bilayers controls their biological function [1]. In man-made colloidal suspensions, colloidal particles can be confined to diffuse in a plane by walls [2] or electrostatic forces [3]. While much is understood about complex fluid-fluid interfaces, fundamental questions regarding the diffusive transport at interfaces remain unanswered, and while the diffusion of colloids and polymers on a fluid-fluid interface has been studied theoretically since the 1970s [4], *collective diffusion* in a monolayer of colloidal particles confined to a fluid-fluid interface has only recently been explored in some detail [5].

We developed an efficient algorithm for Brownian dynamics with hydrodynamic interactions, suitable for modeling diffusion of spherical colloids of hydrodynamic radius a confined to a two-dimensional plane. We used this algorithm to perform large-scale particle simulations and studied collective diffusion on fluid-fluid interfaces (Quasi2D) and in two-dimensional liquids (True2D). We also perform a new type of numerical experiment by coloring (labeling) a subset of the particles and studying the total and color density fluctuations as necessary to model experiments based on fluorescent techniques.

The nonzero compressibility of the three dimensional flow at the fluid-fluid interface leads to a nonzero divergence of the mobility matrix. Under the action of hydrodynamic fluctuations in the fluid, this compressibility acts like a pairwise repulsive potential of order $k_B T(a/r)$, and changes the nature of diffusion dramatically (see Fig. 1).

The thermal origin of this repulsion demonstrates that density functional theories that do not account for thermal fluctuations are incomplete even for ideal systems. We found that the effective particle-particle repulsion leads to a nontrivial reduction of the long-time self-diffusion coefficient as the packing density increases, even for an ideal gas of non-interacting particles (Fig. 2).

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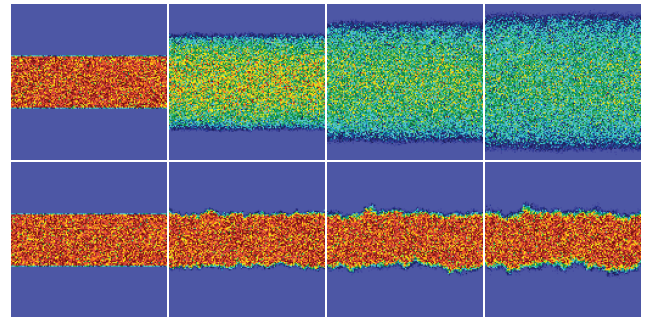


Fig. 1. Diffusion of a density perturbation initially localized in the middle third of the domain. We show snapshots at several points of equal relative time for BD-q2D (top row) and BD-t2D (bottom row). The images show the number density computed by counting the number of particles in each cell of a 128x128 grid; the color bar goes from 0 (blue) to 0.4 (red).

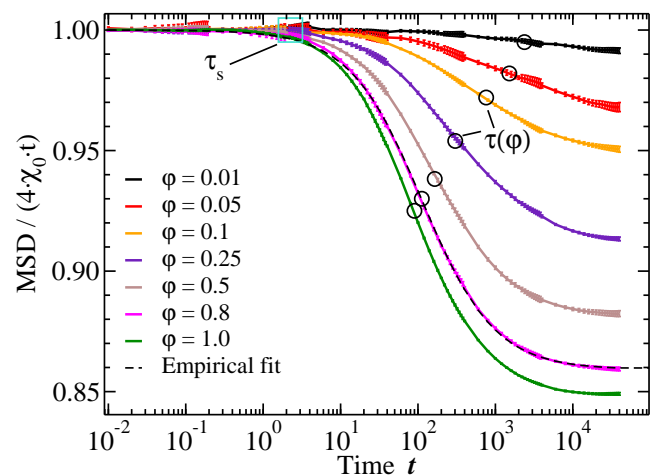


Fig. 2. Mean square displacement of a tagged particle in Quasi2D for different packing densities φ (see legend). Black circles denote the cross-over time $\tau(\varphi)$, and the beginning of the subdiffusive regime τ_s is indicated with a cyan square. χ_0 is the short-time self-diffusion.

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