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Hydrodynamics of Colloidal Clustering in Different Dynamic Regimes

P.I. Rafael Delgado Buscalioni, Dept. Física Teorica de la Materia Condensada, Universidad Autónoma de Madrid, Spain; Condensded Matter Institute, IFIMAC

This main global objective of this ACS-PRF-ND9 project was to study the influence of hydrodynamics on the structure and dynamics of very small particles, from dozens of nanometers to micron size. These lengths are the realm of Brownian dynamics, where thermal fluctuations constitute a powerful driving force related to entropy. Before the PRF grant I had experience in macroscale hydrodynamics and on molecular dynamics simulations of different soft matter elements (polymers, membranes…). The PRF grant enabled an important extension in my scientific carreer giving me the possibility of entering into the extremely interesting and impressively broad world of "small particle hydrodynamics". During the PRF grant period I had the opportunity to cover several problems where hydrodynamics plays a relevant role in determining the fate of interacting colloids, polymers and even smaller molecules (lipids). Colloids under the turbulent regime is the second initial objective of the project and it started with quite promising results. However, our collaborator in turbulence had to postpone this project, which we expect to retake as soon as it becomes possible. On the other hand, the PRF grant gave me the possibility of hiring talented young scientist and introduce them in this exciting discipline, which requires expertise not only on hydrodynamics, but also statistical physics and applied maths. In particular, Ming-Yu from China, performed most of the simulations of colloidal gelation, using different colloid models. After his contract funded by the PRF grant, Ming-Yu got a permanent position in China where-from he keeps contact with our group. Using the PRF funds, the master student Raul Pérez (now ph.D student) started a new code to solve the dynamics of many particles in fluid flow. The code, UAMMD, works with graphical processor units (GPU) and runs with amazing efficiency, competing with the best free codes in the web. Using UAMMD, we have then explored interesting problems in many areas: gelation of platelets, polymer dynamics, confined colloidal systems, lipid membranes, plasmonic gold nanoparticles driven by light-matter interactions and the anomalous diffusion of nanoparticles in gels. In the following I will briefly mention these works and comment on the present outcome and expected in near future.

Universally Adaptable Multiscale Molecular Dynamics: UAMMD

UAMMD is a multiscale tool for simulations using particles in different enviroments and with different resolution scales. UAMMD performs molecular dynamics (nanometers), dissipative particle dynamics (hundreds of nanometers), Brownian hydrodynamics (up to tens of microns) and has been recently extended to include immersed boundary Lattice Boltzmann (macroscopic length). Future development will include "smooth particle hydrodynamics" for macro-scale Lagrangian hydrodynamics. UAMMD runs in Graphical Processor Units (GPU's) and compares favorably with HOOMD, which is one of the fastest codes for GPU in the open source "market". UAMMD is adapted to the GPU architecture: the cell-list neighbor search is based on a Z-ordered curve and we have recently included the extremely powerful Positively Split Ewald method [J. Chem. Phys. 146, 124116 (2017)] which adapts the Ewald summation in Fourier space to resolve hydrodynamics in periodic systems. Modules for Monte Carlo simulations (including hard spheres) and for interaction with optic forces have been recently added. UAMMD is publicly available and we acknowledge the PRF grant in its webpage:

https://github.com/RaulPPelaez/UAMMD

Colloidal gelation

The initial target of the present project focused on the problem of aggregation of colloidal particles, and how hydrodynamics might influence its dynamics and final aggregated structures. One of the

main conclusion of this work is that, contrary to what has it been recently published, we do not find evidence that hydrodynamics radically changes the medium size cluster morphologies. Hydrodynamics do modify the rate at which gelation takes place, but it is still not clear to what extent and under what circumstances it changes the final structure. We have submitted a couple of works on this study and expect to become published soon.

Polymer dynamics

Adding hydrodynamic effects in polymer dynamics is certainly a must when one deals with dilute suspensions. The new tools developed during the PRF grant allowed us to investigate the dynamics of star polymers under shear flow. In a series of papers (one of them partially funded by the PRF grant, see below) we analyze the different frequencies appearing in the motion of these stars and correct several misleading concepts in the previous literature on this subject. We consider dilute solutions and polymer melts and, unexpectedly, find that hydrodynamic interactions (induced by polymer-polymer friction forces) is also are also relevant in the viscoelastic dynamics of star polymer melts under flow.

Dynamics of colloids under quasi-2D confination and their fluctuations

In a quasi-2D configuration (q2D) colloidal particles are confined in two-dimensions but embedded in 3D solvent. A confining force is required to keep this confinement, otherwise the entropic force will drive the colloids away from the interface. Precisely, by applying such confining force to the colloids against the osmotic (i.e. entropic) pressure, one immediately transfers momentum from the 3D solvent which spreads over the interface according to the structure of the Oseen tensor. This results in a repulsive drag between the confined colloids which leads to an enhanced collective diffusion. The diffusion coefficient increases strongly with the system's dimension! This phenomena is universal (even in ideal colloids) and it also appears near walls and in air-water interfaces. In a series of works we have i) unveiled the transition from quasi-2D to 3D behaviour, as the confinement is gradually relaxed and ii) investigated the fluctuations of concentration in ideal mixtures under q2D, both under equilibrium and non-equilibrium

Enhancement of the collective diffusion of lipids in membranes

This work explores the consequences of the divergence of collective diffusion for large system's size in the dynamics of lipid membranes. Surprisingly, we find that lipids collective diffusion is enhanced due to a similar mechanism. We have studied this problem using two different methods (molecular dynamics with explicit water, and particles hydrodynamics, using our own codes UAMMD and FLUAM). Both methods, although of quite different nature, produce the same results. Lipids move collectively in the membrane, and their initial diffusion follows the hydrodynamic drag created by the solvent (water), in a way similar to q2D systems. We published recently published a paper in Phys. Rev. Letters on this subject.

Swarm behavior of gold nanoparticles under crossed-laser fields

This research investigates the dynamics of gold nanoparticles under the optic force created by two lasers at right angles and with a quarter-wave lag. As a result from the interaction of hydrodynamic and optic forces in an enviroment with thermal fluctuations, we observe a transition to collective flow with strong nanoparticle currents! This impressive prediction opens new routes to the manipulation of metal nanoparticles by light. Part of this study was performed with the hydrodynamic solver of UAMMD which now also has an optic-module for light-particle interaction. This module solves the optic force arising from the primary laser beam and also from the scattering of light out from each nanoparticle (a many-body effect). This type of optofluidic solver is the first to be developed world-wide. At present we are fostering contacts with experimental groups to reproduce our findings. The manuscript is published in arxiv and it is await for publication in one of the APS journals, Phys. Rev.