EpitopeMap – Exchange Grant Scientific Report

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Project: Brownian motion in an optical trap: theoretical studies and comparison with experiments

The aim of this second visit to the group of Professor Giuseppe Foffi at EPFL (Lausanne) was to continue the theoretical study of the behaviour of a single colloidal particle and its interactions with the environment – typically the solvent in which it is suspended or more generally any object whose size is smaller than that of the particle under observation – started during the previous visit to the group.

A particle which is heavier and bigger than the solvent molecules, but not so big that it is not affected by the stochastic collisions with such smaller molecules, performs a so-called Brownian motion, characterized by random changes in its direction and velocity. The classic theory of Brownian motion relies on the assumption that the fluid exerts on a moving particle a force which is proportional to the velocity of the particle, the proportionality coefficient being the (constant) friction ζ as given by Stokes' Law.

As I mentioned in my previous report, this assumption is correct for macroscopic particles, however when the size is reduced other effects become progressively important. In particular, the forces acting on the particle have to be corrected to include hydrodynamic interactions, consisting in a momentum transfer from the particle to the fluid: the solvent molecules use this momentum to create a vortex around the particle, which in turn affects the speed and direction of its subsequent motion. This introduces persisting correlations, which can be seen in the power-law decay of the long time tail of the velocity autocorrelation function, and has an effect on the friction coefficient and the diffusion constant (Zwanzig & Bixon, 1970).

The problem is solved by describing the solvent via linearized Navier-Stokes equations, while standard equations of motion (with the force and torque exerted by a fluid on the immersed body given in terms of the stress tensor) are written for the Brownian particle, together with boundary conditions at the particle-fluid interface. The degree of compressibility of the fluid and the stickiness of the particle appear as parameters in the derived flow profiles and response functions (Erbas, Podgornik, & Netz, 2010).

Theoretical predictions which are not obtained for the particular case of an incompressible fluid are obviously essential if one wants to describe the behaviour of systems in which compressibility effects are relevant. One such example is the study of the backtracking of a spherical particle slowing down in a compressible fluid: under certain conditions, the velocity of a particle initially set in motion by a sudden impulse has been observed to change sign, which means a reversal of the direction of motion (Felderhof, 2005). This is a direct consequence of hydrodynamic interactions: as the sphere slows down, it generates sound waves in the liquid, which in turn affect its later motion through a reactive force.

My host group chose to use this particular effect in order to check how accurately the computational method they use to simulate fluids (multiparticle collision dynamics (Padding & Louis, 2006)) is able to reproduce the behaviour of a real fluid. I therefore studied the phenomenon from a theoretical point of view to determine under which conditions the effect is expected to be observed.

To do so, I studied the velocity autocorrelation function (VACF) of a spherical Brownian particle, since a reversal of the velocity corresponds to an interval in which the VACF assumes negative values. Using the frequency-dependent friction coefficient given by (Erbas, Podgornik, & Netz, 2010), performing an expansion for small times and employing the approximation methods presented in (Schram & Yakimenko, 1998) to invert the Laplace transform of the VACF, I obtained the approximate velocity autocorrelation function in real space. As the presence of an interval in which such function becomes negative depends on its poles and residues, which in general cannot be analytically computed, I performed a numerical analysis of the approximate VACF and obtained the conditions under which backtracking is observed; these depend on the ratios between the relevant time scales (kinematic versus sonic) and densities (particle density versus fluid density) of the system.

Bibliography

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