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Micro and nano fluid mechanics

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ABSTRACT: The current task of contemporary fluid mechanics evidently moves from modeling large scale turbulence to lower, molecular scale limit, where assumption of a continuous and deterministic description becomes questionable again. Once the scaling length of flow becomes comparable with structure dimensions, transport phenomena are strongly modulated by molecular interactions and its proper interpretation needs involvement of deeper physics. New experimental tools largely help in understanding transport phenomena at nanoscales. In the following review we give few examples of problems appealing for new theoretical and numerical models embracing continuous flow modeling with molecular scale phenomena.

1 INTRODUCTION

It is generally understood, that the main difference between macro- and nano-scale mechanics originates from rapidly increasing surface to volume ratio along with the decreasing of object size. A total surface of one nanometer particles filling volume of a cubic centimeter is 6000 square meters! Hence, nanoscience is mainly a science of surface forces and surface interactions. It applies particularly to fluids.

The field of microfluidics is characterized by the study and manipulation of fluids at the submillimeter length scale. The fluid phenomena that dominate liquids at this length scale are measurably different from those that dominate at the macroscale. For example, the relative effect of the force produced by gravity at microscale dimensions is greatly reduced compared to its dominance at the macroscale. Conversely, surface tension and capillary forces are dominant at the microscale. These forces can be used for a variety of tasks, such as passively pumping fluids in microchannels, precisely patterning surfaces with user-defined substrates, filtering various analytes, and forming monodisperse droplets in multiphase fluid streams for a variety of applications (Tabeling 2006). Observing biology one obtains plethora of valuable hints how to proceed with such processes (Sasai 2013). One of the fundamental problems in microfluidics is lack of turbulence. Hence, all involved transport processes are based on laminar, creeping flows with very inefficient at such scales diffusion.

Nanoscale fluid mechanics touches completely new area, rediscovered after biologists. Intercellular and cellular transport of molecules, subcellular organelles, and cells immersed in an aqueous environment is based on "stochastic" transport phenomena. Molecular diffusion becomes very efficient at short distances. Whereas typical biomolecule needs hours to be transported by diffusion for distances of centimeter, such transport takes less than 1 s at subcellular distances. Hence, random walk of nano-objects driven by intermolecular interactions initiates all living processes, whereas hydrodynamic interactions play mainly role of a drag force, modifying species mobility.

Physics of nanoscale mechanics can be more or less completely described using quantum mechanics. The methods, called ab initio, are rigorous but limited by present-day computers to systems containing a few hundred atoms at most. To determine the properties of larger ensembles of atoms the Molecular Dynamics method is commonly used, enabling studies of billions of atoms with effective interatomic potentials. Still its practical applications are limited by time and space scales to first nanoseconds of the analyzed phenomena and several nanometers in space. Hence, modelling fluid flow in micro- and nano-scales needs specific technique, which is based on assumption that fluid particle can be represented as a cluster of atoms. Effective clustering can be built using the so-called Voronoi tessellation, describing a special kind of decomposition of the flow domain (Czerwińska 2004). Such coarse grained modelling is useful for general flow description, but needs predefined interactions if we approach molecular distances to interpret specific phenomenon.

Despite of all technical and physical problems with direct experimental analysis of nano/micro scale flows, physical experiment appears to be the only way to validate simplified assumptions, which by definition have to be incorporated to theoretical

models. Some experimental examples refer to observing the mother nature, some of them have been recently proposed using completely new for fluid mechanics techniques, like fluorescent microscopy, Atomic Force Microscopy (AFM), and Optical Tweezers (OT).

Most of microfluidic problems concern multiphase flow, suspensions of micro and nanoparticles, cells or macromolecules (proteins, DNA). Understanding and properly interpreting fluid-particle interaction is crucial for interrogating such systems. One of the valuable optical tools is based on Brownian motion (Breuer 2004). The local and bulk mechanical properties of a complex fluid can be obtained by analyzing thermal fluctuations of probe particles embedded within it.

Thermal fluctuations generated by molecules are not only noises, it has been demonstrated that such fluctuations are fundamental to the function of biological systems. Several results are possible with the same probability, in contrast to a mechanical system in which the result is deterministic. In ensemble measurements, the obtained values, which are average values over many molecules, have been usually wrongly interpreted as deterministic values. However, in biological systems, the average values are not necessarily effective, but the values of individual molecules play a decisive role (Kochańczyk et al. 2013, Jaruszewicz et al. 2013).

The biomolecular system may spontaneously fluctuate, and one of the two states occurs alternately. Preferential binding of ligands to one of the spontaneously fluctuating structures of proteins leads to their activation or deactivation. This mechanism appears essential for a long scale evolutionary development of leaving species, and at short time scale to create signaling paths for early immune response of individual cells (Tay et al. 2010). Hence, looking at the "bottom" of our fluid mechanics, there is no place for steady, unique and predictable modeling. Rather, by analogy to quantum mechanics, we have to talk about the most probably evolution of the analyzed system. As an illustration of the difficulties, in the following we cope with two intriguing problems, kinematic boundary conditions in micro and nano scales flow, and mobility of nano-objects suspended in liquids.

2 TO SLIP OR NOT TO SLIP

One of the primary questions, which appeared when fluid mechanists started to play with microfluidics, concerned the interactions between liquids and solid surfaces. From the physical point of view it seems obvious that molecules of liquid cannot be arrested at the solid surface, otherwise the local thermodynamic parameters of liquid should abruptly change. This problem is of fundamental physical interest and has practical consequences in rarefied gas flows. Recently it became rediscovered for small-scale systems, including transport phenomena in biological fluids (Lauga et al. 2005).

The physics of hydrodynamic slip may have different origins. Purely molecular slip is clearly relevant in case of gases. For very small channels formed by Carbon NanoTubes (CNTs) possible drag reduction is expected due to the synchronized slippage of water molecules. It is interrelated with molecular structure of water. For constrained conditions, i.e. tube diameter below 1 nm, water forms a long single molecular chain (Thomas et al. 2009). Thus, its transport properties largely deviate from continuous understanding of fluid flow.

For dense fluids several additional factors appear to play a more or less significant role. One of them involves wetting properties of the solid surface. Molecular scale roughness allows for creation and stabilization of nanobubbles. Such trapped on the wall nanobubbles may effectively work as a gaseous slip layer, responsible for super-hydrophobic surface properties (Borkent et al. 2007).

Identification of the slip appears not a simple task, both numerically as experimentally. Molecular simulations performed up to now neither confirm nor exclude possible deviation from the classical "non-slip" condition formulated by Navier (Navier 1823). Using continuum mechanics we have to combine a grid size that copes with a nanometer while covering enough space to also include a millimeter scale flow. Experimentally, our techniques for looking at the very small objects (e.g., atomic force microscopy) are slow and cannot cope with large areas very well. The attempts to look at both these scales simultaneously show how our intuition about the relationships between nanoscale objects and macroscopic objects can fail badly.

Classical microscopy used for nanoscale observation has resolution limited by the light wavelength of about 500 nm micrometers. Evaluating diffraction disks the measured position of particle coordinates in plane perpendicular to the optical axis can be improved by order of magnitude. However, resolution in depth, along optical axis, remains very low (tenths of micrometer), and is defined solely by focal depth of the microscope lens.

Total Internal Reflection Fluorescence microscopy (TIRF) helps to bypass some of these limitations offering a possibility to locate objects position with resolution of about 20 nm. Laser light illuminating object undergoes total internal reflection at an interface between investigated medium (liquid) and the wall (glass), and part of the light penetrates into the medium parallel to the interface with an intensity that decays exponentially with the normal distance from the interface. This evanescent wave

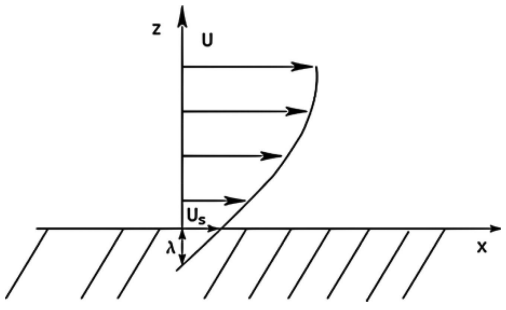

Figure 1. Schematic definition of the slip velocity U_s and the slip length λ for the fluid flow over solid wall.

illumination has been used extensively in the life sciences. Recently it was rediscovered in microfluidics for near wall flow measurements. The main advantage of the method is possibility to reduce the depth of focus of the acquisition system. Hence, it became possible to obtain images of particles, which are in the direct vicinity of the wall.

In our recent study of the Brownian motion of fluorescent particles observed close to the wall (Zembrzycki et al. 2012), the deviation of the particle diffusion rate has been interpreted as an evidence of the slip boundary conditions. According to the theoretical model by Lauga & Squires (2005) the diffusion coefficient of a single colloidal nanoparticle is directly related to the distance from the wall, and the slip velocity. We applied this prediction to determine the slip length from measured and calculated variations of the diffusion coefficient of particles as a function of distance from the wall (Zembrzycki et al. 2012). For this purpose the effect of the wall on the Brownian motion of nanoparticles suspended in water was examined experimentally and compared with numerical simulations performed by Molecular Dynamics approach. The outcome is in the range of uncertainty found in the literature. For a relatively large nanoparticle used in the experiments (300 nm diameter) the evaluated slip length (Fig. 1) measured at 170 nm from the wall appears to be nearly 300 nm. In the numerical analysis, for much smaller particles (24 nm) the evaluated slip length is less than 4 nm. Our difficulties with proper interpretation of available measurements of slip length performed by particle tracking became partly understandable if we look at inherent factors modifying mobility of nanoobjects. We discuss it in the following chapter.

3 MOBILITY OF NANOPARTICLES

Micro and nano scale motion is coupled or sometimes mainly driven by molecular diffusion, direct effect of molecular structure of our environment. Diffusion governed by Brownian motion is an efficient transport mechanism on short time and length scales. Even a highly organized system like a living cell relies on the random Brownian motion of its constituents to fulfill complex functions. A Brownian particle will rapidly explore a heterogeneous environment that in turn strongly alters its trajectory. Thus, detailed information about the environment can be gained by analyzing the particle trajectory. For such an analysis spatial resolution down to the nanometer scale is needed. High resolution is directly connected to the requirement to observe the motion on short time scales. However, at short time scales, the inertia of the particle and of the surrounding fluid can no longer be neglected, and one expects to see a transition from purely diffusive to ballistic motion (Huang et al. 2011). The effect is not negligible for transport phenomena observed in nanoscales, e.g. single-molecule reactions, which are basis for transcription of encoded in DNA information. Thus, for its complete understanding an analysis of Brownian motion at very short time scales is necessary, taking effects of inertia into account.

Biochemical reactions in living systems occur in media of very high molecular concentration. In fact it is difficult to talk about diffusion of molecules at such crowded environment, the interactions between macromolecules hinder their displacements, limiting transport and signaling functions (Długosz & Trylska 2011).

Observation of Brownian motion of microobjects is a classical basis for particles size measurements, evaluation of liquid properties (viscosity, microrheology), analyzing particle-wall interactions, and many others. Nevertheless even such a seemingly simple problem creates plethora of uncertainties. In all applications it is necessary to maintain the colloid well dispersed and to avoid the formation of aggregates (Chassange & Ibanez 2014). Moreover, it is absolutely necessary to know the fluid-solid interaction in nanoscale and the hydrodynamic properties of the particles.

The equilibrium state and the hydrodynamic properties of many colloid systems in aqueous medium is affected by several environmental parameters. The ionic character of water solutions needs, besides analysis of hydrodynamic friction (famous Stokes formulae), evaluation of surface—liquid interactions. The evaluation of surface charges, ionic streams, creation of the electrostatic double layer theoretically is possible with help of Derjaguin-Landau-Verwey-Overbeek (DLVO) theory (Israelachvili 2011). In practice, we are far from incorporating all necessary molecular and ionic interaction to our macro hydrodynamics, hence commonly used empirical expression called "hydrodynamic diameter" effectively

shadows our lack of knowledge. In several cases such simplification can be sufficient for chemical engineering; it becomes unacceptable if size of colloid particles strongly decreases. Any ionic layer, streams of ions attaching particle, steric interactions with suspended molecules, effectively decrease nanoparticle mobility. A proper prediction of such effects is crucial for understanding transport processes at the single cell level.

Figure 2 illustrates our attempts to evaluate size effect for Brownian nano particles (Pawłowska et al. 2014). It is evident that decreasing particle size, the effective (hydrodynamic) diameter affects their diffusion. Moreover, variation of ionic strength of the liquid environment (Fig. 3) strongly modifies diffusion of nanoparticle additionally changing its hydrodynamic diameter.

Detailed experimental analysis of interactions of liquid molecules and surface molecules of individual particle is very difficult. Hence, in practice more or less sophisticated hydrodynamic models are implemented to interpret observed variation of the apparent particle diameter (in fact friction coefficient). Such models used later for measuring and sorting macromolecules are in common use, despite questionable theoretical background given by fluid mechanists.

Recently, a new optical tool called Optical Tweezers (OT) expanded our traditional instrumentation, creating possibility for undisturbed measurements of forces and position in picoNewton and nanometer scales. Detailed evaluation of the forces responsible for particle—particle interactions became possible with help of combined AFM-OT apparatus developed at IPPT (Pierini et al. 2015).

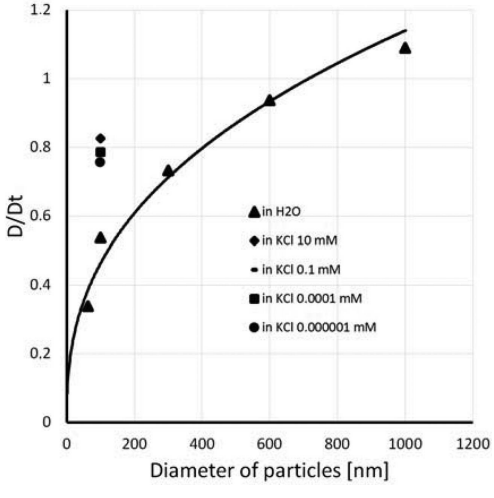

Figure 2. Relative diffusion coefficient measured for polystyrene nano spheres suspended in water; effect of diameter and solution concentration (100 nm particles).

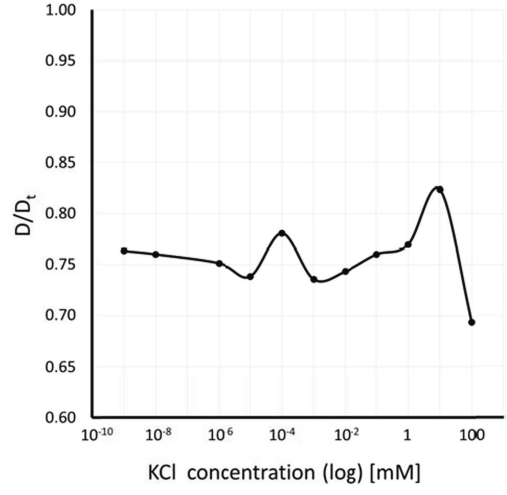

Figure 3. Ionic modulation of diffusion coefficient observed for 100 nm polystyrene spheres suspended in diluted KCl solutions.

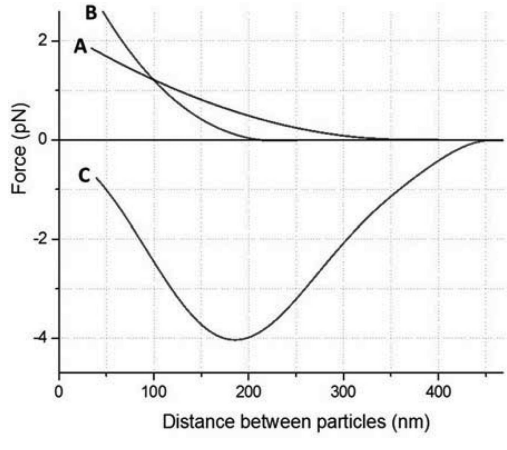

Figure 4. Interaction of two individual colloid particles measured by combined AFM—Optical Tweezer apparatus (Pierini et al. 2015). Data collected for two KCl solutions: 10 3 M (A), 10 5 M (B), and for pure water (C).

Approaching two nanoparticles at nanoscale distances and evaluating intermolecular forces for picoNewton range (Fig. 4) opens attractive possibility to validate existing models of ionic and steric interactions in colloidal suspensions. Dragging, towing single particle trapped by OT allows to perform precise analysis of forces involved by liquid environment, wall interactions, and particle-particle interactions.

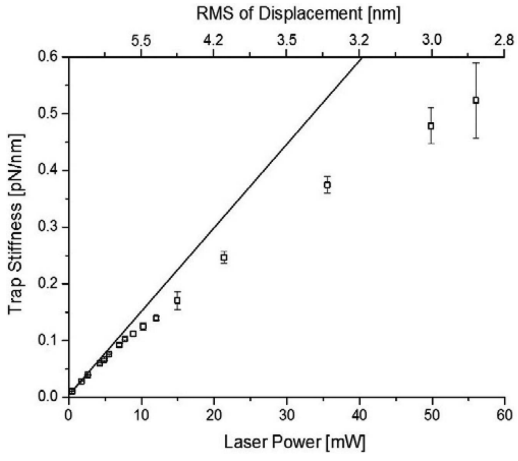

Figure 5. Stiffness of the Optical Tweezers evaluated for 1 μm polystyrene particle suspended in water; particle diffusion increases as the mean Brownian displacement decreases (upper axis). Straight line—large displacements theoretical limit.

An electronic way of signal analysis allows for thermal motion of particle trapped by OT to be evaluated with MHz sampling frequency and displacements below 1 nanometer. Hence, one of the fundamental problems of single particle mobility, namely ballistic regime and effects of inertia creating time dependent recirculation of surrounding liquid molecules, could be proven using OT (Franosch et al. 2011).

In our preliminary study OT developed at IPPT have been used to analyze Brownian motion of trapped polystyrene particle (Zembrzycki et al. 2014). It appears that already at sampling times of 10 kHz diffusion becomes influenced by ballistic regime of molecular interactions (Fig. 5).

4 WORM-LIKE CHAIN (WLC)

The flow of deformable objects (fibers, polymer chains) has a non-Newtonian character, strongly influencing its short time response at microscopic level (Gittes et al. 1993, Jendrejack et al. 2004).

Under the flow these objects are oriented, deformed, and coiled leading to a macroscopic variation of the transport properties.

The flexibility of long biopolymers reflects important aspects of their biological functions. The bending capabilities of DNA have special meaning if we realize that nearly 2 m long human DNA has to be transported through the cell membrane and finally packed into the cell nucleus. The elasticity of cytoskeleton constituents such as actin filaments, microtubules determines cell shape and mechanical response.

The microscopic structures, as well as the macroscopic response of flexible filaments, depend on both the nature of the suspended objects and the flow configuration. Linking mechanical and microscopic properties of the suspended objects to the macroscopic response of the suspension is one of the fundamental scientific challenges of soft matter physics and remains unsolved for a large number of situations typical for intercellular transport of proteins and ligands.

Most of the biomolecules have strongly elongated form, far from idealistic ball like shape. Their penetration through the crowded cellular environment is strongly enhanced by its shape flexibility. The interplay between crowding and thermal bending allows for their controlled mobility.

Deformations of long micro and nano objects are induced not only by thermal fluctuation (Liu et al. 2004), but they are also introduced by non-linear hydrodynamic interactions, leading to some intriguing behavior. For example, it has been predicted that long fibers may perform spectacular windings to form more or less stable knots, phenomenon of fundamental importance for biological macromolecules (Sadlej et al. 2010, Kuei et al. 2015).

Despite recent advances in this field, there is still lack of experimental investigations to validate assumptions of the theoretical and numerical models. Deficiency of experimental studies is mainly due to the absence of good experimental model systems that allow for determining and controlling elasticity and geometry of analyzed flexible objects.

Observation of proteins or DNA is still mostly qualitative, limits of the optical methods permit to find out some predicted characteristics only. Therefore, to systematically investigate the interactions of long molecules with given flow and the resulting macroscopic transport properties, synthetic experimental models of flexible objects can be useful. Hence, we aimed to produce flexible nanofibres to mimic behavior of "worm-like chains". Alas, winding of the objects predicted by simple Stokesian model could not be confirmed in the experiment (Sadlej et al. 2010). It was probably due too high stiffness of nanofibres used in our experiments.

Recently, we have developed new core-shell electrospinning procedure allowing for constructing highly deformable microscopic filaments (Fig. 6), with typical diameter of 100 nm and contour length ranging from single micrometers to millimeters (Nakielski et al. 2015). Introducing such objects directly into microfluidic channels allowed us to observe their deformations due to the flow (Fig. 8) as well as those induced by thermal fluctuations (Table 1). It can be seen that even short

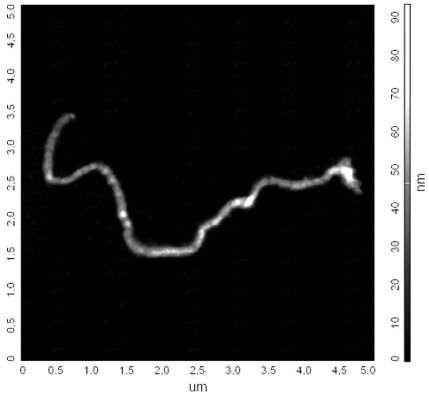

Figure 6. Hydrogel nano-filament observed under an atomic force microscope; contour length 7 μm, diameter 80 nm.

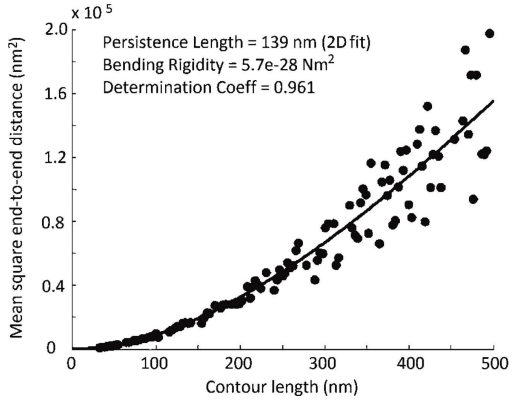

Figure 7. Mean square end to end distance to calculate persistence length of 48 kbp λ-DNA observed with atomic force microscope on mica plate.

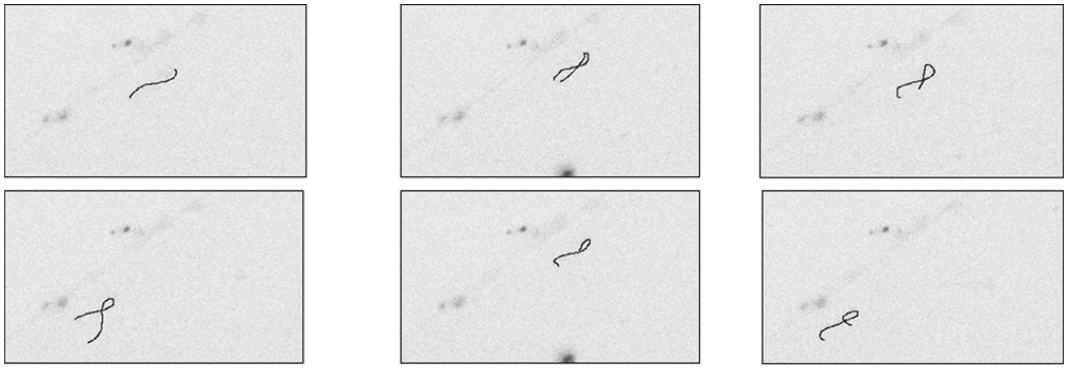

Figure 8. Single highly flexible nanofilament conveyed by Poiseuille flow in 100 μm microchannel. Filament diameter 100 nm, contour length 25 μm. Flow Reynolds number based on the microchannel size equals 4.10–3. Sequence of images (not ordered in time) extracted from the movie recorded under fluorescent microscope and displayed as negatives; image width ~50 μm. There are visible effects of 2-D projection which apparently change contour length of the filament. By selecting images preserving contour length we are able to limit our analysis to in-plane filament deformations.

pieces of filaments exhibit complex mobility, hardly described by the simple theoretical model with translational and rotational diffusion coefficients of prolate ellipsoid (Ortega & Torre 2003).

Additional degrees of freedom created by shape deformation, coiling and uncoiling of longer filaments, change overall redistribution of thermal energy and effective mobility of such objects. The last effect has additional advantage; analysis of thermal fluctuations of flexible objects can be used to evaluate their persistence length (Kratky & Porod 1949), directly correlated with its mechanical properties. Formally the persistence length of long polymers reflects their bending rigidity, parameter describing distance over which the chain of molecules "remembers" its initial position.

For most of biomolecules persistence length varies from nanometers to millimeters (Gittes et al. 1993). Typical values of persistence length obtained for our hydrogel filaments range from 5 μm to 500 μm (Nakielski et al. 2015), being very close to relative values measured for DNA chains (Fig. 7).

Direct observation of shape fluctuations for molecules is presently prohibited by long time illumination necessary to visualize nano objects under optical microscope. Hence, such analysis of DNA persistence length is performed for objects collected onto a functionalized mica substrate, where object-wall interactions may strongly bias statistics of observed deformation variants. In that case collected AFM images of molecules (DNA) are segmented to evaluate local bending curvature (so

Table 1. Translational and rotational diffusion coefficients evaluated from Brownian motion of typical samples of nanofilaments. Experimental data are compared with theoretical diffusion coefficients given for the ideal non-deformable, prolate ellipsoid. Diffusion was measured between two cover slides, about 50 μm apart. Measurements performed in water at temperature T = 30°C, viscosity ηs = 1.0210–3 Pa s, L—filament contour length, R—radius.

	L/2R	D_a (μm²/s)	D_b (μm²/s)	D_r (rad²/s)	L (μm)	Shape
Exper.	43.0	0.052	0.035	0.0006	21.5	
Theor.	43.0	0.144	0.090	0.0019		
Exper.	31.2	0.093	0.067	0.0025	15.6	
Theor.	31.2	0.182	0.116	0.0045		
Exper.	29.8	0.059	0.140	0.0029	14.9	
Theor.	29.8	0.189	0.121	0.0051		
Exper.	47.8	0.231	0.053	0.0005	23.9	
Theor.	47.8	0.133	0.083	0.0014		
Exper.	31.4	0.266	0.070	0.0023	15.6	
Theor.	31.4	0.182	0.116	0.0044		
Exper.	48.4	0.112	0.105	0.0026	24.2	
Theor.	48.4	0.132	0.082	0.0013		
Exper.	104.8	0.054	0.070	0.0005	52.4	
Theor.	104.8	0.072	0.044	0.0002		
Exper.	171.0	0.039	0.150	0.00005	85.5	
Theor.	171.0	0.049	0.029	0.00004		
Exper.	105.6	0.029	0.087	0.0002	52.8	
Theor.	105.6	0.072	0.043	0.0002		

called cosine method), or simply end-to-end length (Fig. 7) is used to build statistical description of molecule deformation.

It is worth to underline that our observations of thermal fluctuations of filaments take place for objects freely suspended in liquid. Hence, despite of 2-D projection limits, we are able to collect statistically relevant sets of data for their thermal deformations. Henceforth, we are able to validate worm-like chain models with material parameterization obtained from the experiment (persistence length). This approach could in the future be used to gain further fundamental understanding of filaments dynamics under flow as a function of their complex mechanical properties as anisotropy or deformability.

It is interesting to note that even for relatively low flow Reynolds number (Fig. 8) we observed typical coiling—uncoiling sequences. It is remarkable similar to WLC modelling performed with Stokesian approach (Sadlej et al. 2010, Kuei et al. 2015). Understanding the link between the microscopic structure of the filaments and the macroscopic flow properties opens the possibility to design nano-objects transported by body fluids for targeted drug release or local tissue regeneration.

5 CONCLUSIONS

Recent development of experimental techniques applicable to fluid mechanics of micro and nano scale permits to have a closer look at applicability of existing mechanical models to small scale phenomena. In the following we have presented few selected problems characterizing nano and micro scale fluid dynamics, namely kinematic boundary condition at solid interfaces and suspension of nano scale intrusions, like nanoparticles and long, deformable filaments. Evidently, similarly to near field fluctuational electrodynamics (Song et al. 2015), *fluctuational fluid mechanics* is needed to describe transport phenomena at nanoscales.

In computational modeling of nanoscale flow problems, similarly to solid mechanics science, there is still unsolved problem of merging atomistic scales with nano, micro and macro systems. In case of fluid mechanics there is additional difficulty. Merging of time stepping for all scales has to be performed for very short time scales. This is challenging, still not available approach. Therefore, we have to cope with simplified models, where solely experimental validation may offer background for tuning and adjustment of crucial model parameters.

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