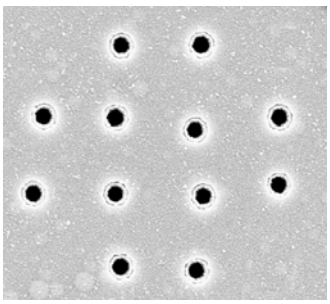
Swiss Soft  Days

2nd WORKSHOP
Lausanne 23.06.2010

Sponsored by

ÉCOLE POLYTECHNIQUE
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Local Organizer

Suliana Manley
EPFL
Laboratoire de biophysique
expérimentale

Giuseppe Foffi
EPFL
Groupe Foffi (GR-FO)

Program

10.00-10.05	Welcome	
Theme 1: Materials & Assemblies		
10.05-10.20	Dietsch (U Fribourg)	Nanoengineering of responsive hybrid anisotropic particles
10.20-10.35	Sanchez-Ferrer (ETHZ)	Inorganic-organic elastomer nanocomposites from nanoparticles as crosslinking agents
10.35-10.50	Balog (PSI)	Correlation between morphology, water uptake, and proton conductivity in radiation grafted proton exchange membranes
10.50-11.05	Limbach (Nestlé)	Simulation of small molecules at the ice solution interface
11.05-11.20	Bischofberger (U Fribourg)	Bulk structure of water determining the hydrophobic hydration of polymeric amphiphiles
11.20-11.35	Wolf (IBM)	Self-assembly and soft lithography for nanostructure fabrication
Theme 2: BioPhysics		
11.35-11.50	Radenovic (EPFL)	DNA/ graphene hybrid nanostructures
11.50-12.05	Lim (U Basel)	Synthetic protein targeting to polyethylene glycol with associated antibodies
12.05-13.30	**Lunch / Coffee / Posters / Discussion**	
13.30-13.45	Voets (U Fribourg)	Solvent-induced denaturation, self-assembly, and gelation of globular proteins
13.45-14.00	Gabella (EPFL)	Plasma membrane tension and actin-dependent protrusion forces in migrating cells
14.00-14.15	Savin (ETHZ)	Capillary flow occlusion caused by impaired red blood cells
14.15-14.30	Melchionna (EPFL)	Multi-scale modeling of blood flows in extended coronary arteries
14.30-14.45	Pfohl (U Basel)	In situ dynamics and fluidics of soft and biological matter
14.45-15.45	**Coffee / Posters / Discussion**	
Theme 3: Colloids & Polymers		
15.45-16.00	Celebrano (ETHZ)	High sensitivity nanoscopy via interferometric scattering microscopy
16.00-16.15	Krishnan (ETHZ)	Geometry-induced trapping, levitation and assembly of nanometric objects in a fluid
16.15-16.30	Mokhtari (U Fribourg)	Diffusion dynamics of colloidal rods at liquid (solid) / liquid interfaces
16.30-16.45	Sinha (U Geneva)	Prediction of the attractive double layer forces between two different surfaces using charge regulation effects
16.45-17.00	Dorsaz (EPFL)	Diffusion-limited reactions in crowded environments

Abstracts Talks

Theme 1: Materials & Assembly 10.05 – 11.35

Nanoengineering of responsive hybrid anisotropic particles

H. Dietsch

Adolphe Merkle Institute and Fribourg Center for Nanomaterials, University of Fribourg

In the process of creating new model colloidal systems, the control of the morphology of particles is often a challenge. The interest in various anisotropic particles has been recently growing, in particular since it opens new possibilities for optical devices and mechanical reinforcement of polymeric materials. Among the various geometrically anisotropic particles, ellipsoid colloids are especially attractive, since from a fundamental point of view they exhibit both higher packing densities than spheres. One of the limits of ellipsoidal particles is a synthetic issue. In the present paper, I will present an overview of the techniques we use leading to spindle and ellipsoidal shaped particles. Techniques presented can lead to multiresponsive colloidal systems using preferable crystalline growth, nanotemplating or mechanical stretching methods for example.

Inorganic-Organic Elastomer Nanocomposites from Nanoparticles as Crosslinking Agents

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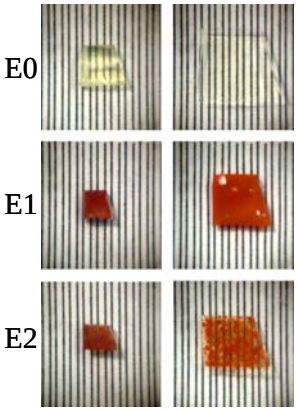
b) University of Fribourg, Adolphe Merkle Institute and Fribourg Center for Nanomaterials.

Magnetic inorganic-organic network nanocomposites are covalently crosslinked polymeric systems which can be found either in swollen state or in dry state and exhibit magnetic nanoparticles dispersed into the polymer matrix.[1] These materials have the combined advantages of organic polymers (flexibility and processability), and the inorganic nanoparticles (magnetic, optical and electrical properties).[2] Consequently they can elongate and contract under magnetic field gradients and behave as soft actuators.[3] The concentration of magnetite particles and the crosslinking density play an essential and important role in their magneto-elastic behavior. These nanocomposites can be formed by the precipitation of magnetic nanoparticles in the organic material before, during, or after the crosslinking process.[4,5]

We report on the synthesis of nanocomposite with integrated ellipsoidal silica-coated hematite (SCH) spindle type particles, which can act as crosslinking agents within an elastomeric matrix. Influence of the surface chemistry of the hematite leading either to dispersed particles or crosslinked particles to the elastomer matrix was studied via swelling, scattering and microscopy experiments. It appeared that without surface modification the SCH particle aggregate and act as defects whereas the surface modified SCH particles increase the crosslinking density and thus reduce the swelling properties of the nanocomposite in a good solvent. For the first time, inorganic SCH particles can be



easily dispersed into a polymer network avoiding aggregation and enhancing the properties of the resulting inorganic-organic elastomer nanocomposite (IOEN).

The unswollen and swollen dimensions for the pure network (E0), with crosslinkable (E1), and non-crosslinkable nano-particles (E2).

1) Zrínyi, M. *Trends Polym. Sci.* **1997**, *5*, 280-285.
2) Althues, H.; Henle, J.; Kaskel, S. *Chem. Soc. Rev.* **2007**, 36, 1454–1465.
3) Zrínyi, M.; Barsi, L.; Büki, A. *Polym. Gels Networks* **1997**, *5*, 415-427.
4) Zrínyi, M.; Barsi, L.; Szabó, D.; Kilian, H.G. *J. Chem. Phys.* **1997**, *106*, 5685-5693.
5) Mitsumata, T.; Ikeda, K.; Gong, J.P.; Osada, Y.; Szabó, D.; Zrínyi, M. *J. Appl. Phys.* **1999**, *85*, 8451-8456.

Correlation between Morphology, Water Uptake, and Proton Conductivity in Radiation Grafted Proton Exchange Membranes

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b) Adolphe Merkle Institute, University of Fribourg
c) Department of Natural Sciences, University of Copenhagen, Frederiksberg
d) Electrochemistry Laboratory, Paul Scherrer Institut

We present small-angle neutron scattering studies of fully hydrated proton exchange membranes. To understand the relationship between morphology, water uptake, and proton conductivity, we applied the technique of contrast variation. Our membranes were synthesized by radiation-induced grafting of poly(ethylene-alt-tetrafluoroethylene) (ETFE) with styrene in the presence of crosslinker (divinylbenzene, DVB), and by sulfonating the polystyrene subsequently. Crosslinking allows the adjustment of the structural density of the grafted component, which highly influences the fuel cell relevant properties, such as water uptake and proton conductivity. We report on the domain structure, as observed by SANS, focusing on the impact of crosslinking on the relationship between the domain structure, water uptake, and proton conductivity.

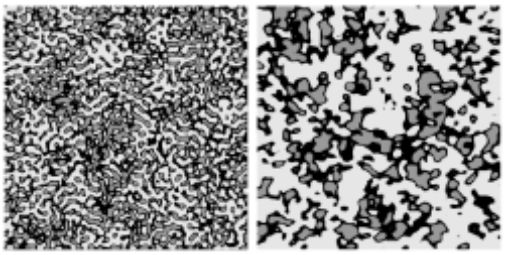

Figure 1: The cross-sectional morphology visualization of a crosslinked and an uncrosslinked membrane in the hydrated state. Color code: Grey denotes non-hydrated domains. Black stands for poorly hydrated domains. The lightest shade marks water rich domains.

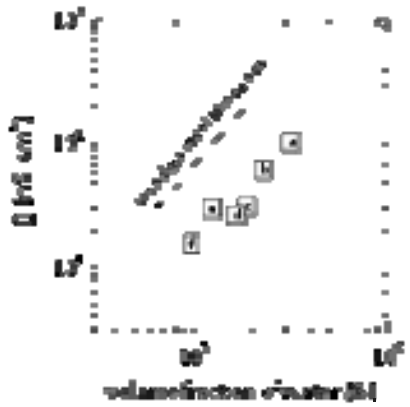

Figure 2: Proton conductivity versus volume fraction of water in the swollen membranes. The crosslinker level is a.) 0%, b.) 5%, c.) 8%, d.) 10%, e.) 15%, f.) 20%.

1) S. A. Gürsel, L. Gubler, B. Gupta, and G. G. Scherer. Radiation Grafted Membranes. In Fuel Cells I, volume 215 of Advances in Polymer Science, pages 157-217. 2008.
2) L. Gubler and G. G. Scherer. Radiation-grafted proton conducting membranes. In H. Yokokawa W. Vielstich, H. A. Gasteiger, editor, Handbook of Fuel Cells: Advances in Electrocatalysis, Materials, Diagnostics and Durability, volume 5 & 6. John Wiley & Sons Ltd, 2009.

Simulation of small molecules at the ice solution interface

Hans Jörg Limbach

Nestlé Research Center, Lausanne

In a large number of industrial applications as well as in biology and medicine ice-water interfaces are frequently encountered. Despite their importance particularly the interaction of other molecules with this interface is only poorly understood. A better understanding of how small molecules influence the structure of frozen food is our main motivation for this study.
There are two facts that make this interface special and challenging: First, compared to an oil-water or water-air interface the ice-water interface is special, both ice and liquid water are both made from the same molecule, water (even though this is true for any solid liquid phase coexistence). Second, most applications are close to the melting point of ice, therefore the interface is highly dynamic and not clearly defined. Due to both reasons it is quite difficult to characterize the interface experimentally
Since both ice and liquid water are hydrophilic it is questionable whether the concept of hydrophilicity is appropriate to characterize the interaction of molecules with the ice water interface. Here I try to establish a new concept which ranks small chemical groups according to their affinity to ice, which I call 'pagophilicity' (from Greek: pagos-ice and philos - love). This would be interesting to know, since it would enable us to find molecules that act as a surfactant at the ice water interface.

Bulk structure of water determining the hydrophobic hydration of polymeric amphiphiles

Irmgard Bischofberger[a], Paolo de los Rios[b] and Véronique Trappe[a]

a) Dept. of Physics, University of Fribourg
b) Laboratory of Statistical Biophysics, ITP, EPF Lausanne

Poly-N-isopropyl acrylamide (PNiPAM) is a neutral amphiphilic polymer which exhibits a lower critical solution temperature (LCST). The general understanding of the existence of a LCST is that at low temperatures the hydrophobic part of the molecule is surrounded by a shell of water molecules that form H-bonds that are stronger and longer-lived than the H-bonds of bulk water. The hydration of the hydrophobic part is thus enthalpically favourable. At the LCST the water molecules have enough thermal energy to get released out of this shell, which leads to a gain in entropy of the water and results in the collapse of the polymer.
We investigate the influence of a second small amphiphile, ethanol, on the solubility of PNiPAM and find that small amounts of ethanol lower the LCST. We discuss this phenomenon considering that ethanol enhances the hydrogen bond network of the bulk water; its presence effectively changes the bulk properties and its effect on the solubility of PNiPAM is indirect.

Self-assembly and soft lithography for nanostructure fabrication

Heiko Wolf

IBM Research GmbH, Zürich Research Laboratory

The special properties of nanoscale objects, such as metal and semiconductor nanoparticles, nanowires, or nanotubes, make them promising building blocks of novel optical and electronic devices. The systematic fabrication of devices usually requires the integration of these heterogeneous materials into a larger, ordered structure. More complex device architectures may, in addition, entail the selective assembly of several different types of small objects into pre-defined locations or demand an assembly with the correct orientation when such objects are non-spherical. We explore a technique that uses a combination of templated self-assembly and printing with elastomeric stamps to fabricate defined particle arrays with high yield and accuracy. The assembly step is performed from an aqueous colloidal suspension onto a patterned template. The template is a silicone elastomer replica of a 3D-structured silicon master. During an adhesive transfer step, the assembled particles are printed onto the target substrate.

Theme 2: BioPhysics 11.35-14.45

DNA/ Graphene hybrid nanostructures

Aleksandra Radenovic, Sudhir Husale, Floriano Traversi and Andras Kis

Laboratory of nanoscale biology, Institute of Bio-engineering, EPF Lausanne

Graphene is a single layer of two-dimensional sp^2 carbon atoms that are closely packed into honeycomb two-dimensional structure. This new kind of carbon nanostructured material was first produced in 2004 [1] and since then it has attracted great attention due to its exceptional electronic [2-3], mechanical [4-7] and thermal properties[8]. Those properties make graphene an ideal candidate for the future substrate of hybrid organic-inorganic nanostructures. On the other hand, hybrid organic–inorganic nanostructures are appealing due to their ability to combine the best properties of the two components. Among organic molecules, DNA properties such as hybridization, convenient synthesis of designed sequences, and its widespread application in structural DNA nanotechnology [9-11] could match exceptional graphene properties. With a new naonmaterial such as graphene, come new opportunities to revisit old problems as well as to pose new ones. Indeed, ssDNA-carbon-nanotube (ssDNA-CNT) hybrid nanostructures have been used for following applications: CNT solubilization [12], sorting [13] and patterned placement of nanotubes [14]Our group works in the field of DNA nanotechnology based research where our main focus is to implement DNA origami structures into nanoelectronics and use various nanostructures for biological applications. DNA origami is a technique in which a long single strand of DNA is folded into any predetermined shape and can display 6-nm-resolution patterns of binding sites, thus can allow complex arrangements of carbon nanotubes, silicon nanowires, or quantum dots. We use graphene as substrate for controlled placement and orientation of DNA origami in a desired direction.

1) K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva and A. A. Firsov, Science **306** (5296), 666-669 (2004).
2) K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos and A. A. Firsov, Nature **438** (7065), 197-200 (2005).
3) Y. B. Zhang, Y. W. Tan, H. L. Stormer and P. Kim, Nature **438** (7065), 201-204 (2005).
4) S. S. Verbridge, H. G. Craighead and J. M. Parpia, Applied Physics Letters **92** (1), -(2008).
5) J. C. Meyer, A. K. Geim, M. I. Katsnelson, K. S. Novoselov, T. J. Booth and S. Roth, Nature **446** (7131), 60-63 (2007).
6) J. C. Meyer, A. K. Geim, M. I. Katsnelson, K. S. Novoselov, D. Obergfell, S. Roth, C. Girit and A. Zettl, Solid State Communications **143** (1-2), 101-109 (2007).
7) J. S. Bunch, A. M. van der Zande, S. S. Verbridge, I. W. Frank, D. M. Tanenbaum, J. M. Parpia, H. G. Craighead and P. L. McEuen, Science **315** (5811), 490-493 (2007).
8) A. A. Balandin, S. Ghosh, W. Z. Bao, I. Calizo, D. Teweldebrhan, F. Miao and C. N. Lau, Nano Letters **8** (3), 902-907 (2008).
9) S. M. Douglas, H. Dietz, T. Liedl, B. Hogberg, F. Graf and W. M. Shih, Nature **459** (7245), 414-418 (2009).
10) E. S. Andersen, M. Dong, M. M. Nielsen, K. Jahn, R. Subramani, W. Mamdouh, M. M. Golas, B. Sander, H. Stark, C. L. P. Oliveira, J. S. Pedersen, V. Birkedal, F. Besenbacher, K. V. Gothelf and J. Kjems, Nature **459** (7243), 73-U75 (2009).
11) P. W. K. Rothemund, Nature **440** (7082), 297-302 (2006).
12) M. Zheng, A. Jagota, E. D. Semke, B. A. Diner, R. S. Mclean, S. R. Lustig, R. E. Richardson and N. G. Tassi, Nature Materials **2** (5), 338-342 (2003).
13) M. Zheng, A. Jagota, M. S. Strano, A. P. Santos, P. Barone, S. G. Chou, B. A. Diner, M. S. Dresselhaus, R. S. McLean, G. B. Onoa, G. G. Samsonidze, E. D. Semke, M. Usrey and D. J. Walls, Science **302** (5650), 1545-1548 (2003).

14) R. S. McLean, X. Y. Huang, C. Khripin, A. Jagota and M. Zheng, Nano Letters **6** (1), 55-60 (2006).

Synthetic protein targeting to polyethylene glycol with associated antibodies

Janne T. Hyotyla[a], Jie Deng[b] and **Roderick Y.H. Lim[a]**

a) Biozentrum and the Swiss Nanoscience Institute, University of Basel
b) Institute of Materials Research and Engineering, A*STAR (Agency for Science, Technology and Research), 3 Research Link, Singapore

"Protein targeting" refers to the sorting mechanism by which proteins are delivered to the correct spatial location within the cell. This ability to sift out and direct specific molecules via molecular recognition from a biologically complex environment to site-selective locations with - nanoscale precision - is truly unprecedented from a physical point of view. In this work, we have conceived of a synthetic system that replicates the basic hierarchical principles of protein targeting to sort proteins from a complex liquid environment to highly localized polyethylene glycol (PEG) brushes with nanoscale precision. Owing to its non-fouling property, only specific secondary IgG "cargoes" are exclusively targeted to PEG via the "chaperone"-like action of PEG-binding antibodies whereas unspecific molecules are repelled. Our assay is robust, reversible and reproducible in blood serum, which attests to the unique properties of PEG that complement its effectiveness as a biointerface material (i.e. PEGylation). By harnessing the protein-like behaviour of polymers for bio-molecular recognition, synthetic protein targeting imparts hierarchical control over molecular transport processes in multi-component systems with wide-ranging implications spanning from bio-sensing technologies to micro/nanofluidic transport processes more generally.

Solvent-induced denaturation, self-assembly, and gelation of globular proteins

Ilja Voets[a], Christian Moitzi[a], Elizabeth Areas[b] and Peter Schurtenberger[a]

a) Adolphe Merkle Institute, University of Fribourg
b) Departamento de Química Fundamental, Instituto de Química, Universidade de São Paulo

Understanding and controlling the behaviour of (concentrated) protein solutions is of central importance to various fields of science and industry, ranging from the preparation of highquality crystals for protein crystallography to the prevention of undesirable aggregation in protein condensation diseases and from the stability of the cytosol to the preparation of highprotein foods. We thus need to better understand protein-protein interactions that govern both the equilibrium and non-equilibrium phase behaviour, determining which phase (crystal, fluid, gas) is the most favourable in terms of free energy and whether it is accessible; i.e. whether the system is not kinetically arrested along the path towards thermodynamic equilibrium. The task is far from easy as the total interaction potential consists of many contributions due to electrostatic and van der Waals forces, excluded volume and hydration effects, hydrogen bonding, salt bridging, and ion binding, and so forth. However, it has been successfully demonstrated that - in certain cases - the 'true' interaction potential can be replaced by a much more simplified form consisting for example of a short-range attraction and a long-range repulsion.
In this contribution, we will discuss the effect of dimethylsulfoxide (DMSO) addition to aqueous protein solutions under conditions where the proteins interact through a mixed

potential consisting of a short-range attraction and a long-range repulsion. We are interested in the solvent mixture DMSO/H2O as it has been shown to generate transparent lysozyme gels above certain critical values of both the DMSO volume fraction and lysozyme concentration.
We compare solvent-induced denaturation, self-assembly, and gelation of various globular proteins and attempt to relate the differences in behaviour to differences in the isoelectri points and structure in terms of the amino acid composition, amino acid sequence, and the amount of disulfide bonds.

Figure 1. Artist impression of DMSO-induced denaturation in lysozyme/DMSO/water solutions. The different conformational states are referred to as 'quasi-native' (left), 'unfolded' (middle), and 'partially collapsed' (right).

Plasma membrane tension and actin-dependent protrusion forces in migrating cells

Chiara Gabella[a], Elena Bertseva[b], J. Jacques Meister[a], Ivo Sbalzarini[c], Alexander Verkhovsky[a]

a) Laboratoire de Biophysique Cellulaire, EPF Lausanne
b) Laboratoire de physique de la matière complexe, EPF Lausanne
c) Institute of Theoretical Computer Science and Swiss Institute of Bioinformatics, ETH Zürich

Cytoskeletal forces generated by filament assembly and molecular motors, and plasma membrane tension are believed to control cell shape and participate in cell migration. In particular, membrane tension opposes actin assembly at the leading edge of migrating cells, and it was suggested that the balance between membrane tension and actin assembly force controls edge protrusion rate and cell shape. However, membrane tension and protrusive forces have not been measured under comparable conditions and it is not known if the membrane tension is strong enough to affect the protrusion rate. To answer this question, we used the same optical trap setup to measure membrane tension and protrusive forces in rapidly migrating fish epidermal keratocytes. Membrane tethers were utilized to evaluate membrane tension, while placing the trapped bead in the way of the cellâ€™s leading edge provided the estimation of the protrusion force. The measurements yielded similar ranges of values for both forces (few tens of piconewtons), suggesting that membrane tension can contribute significantly to the control of protrusion rate. Further we investigated if the control by membrane tension contributes to the coordination between front protrusion and retraction at the back of migrating cells. Inhibition of myosin-dependent contractility by blebbistatin resulted in a significant reduction of protrusion rate in migrating keratocytes. However, membrane tension in blebbistatin-treated cells was not higher than in untreated cells, suggesting that a different mechanism rather than a direct control by membrane tension is responsible for reduction of the protrusion rate. We are currently extending the analysis of membrane-cytoskeletal interaction to the protrusion dynamics during injury repair in migrating cells.
Supported by SystemsX IPhD grant.

Capillary flow occlusion caused by impaired red blood cells

T. Savin[a] and L. Mahadevan[b]

a) ETH Zürich
b) Harvard University

The increase of the red blood cells (RBCs') stiffness in the sickle cell disease is believed to be the predominant factor at the onset of vaso-occlusive event. To quantify the effect of cell's deformability on its ability to flow, we studied the motion of a single swollen RBC in a capillary. We used a tapered glass capillary with inner diameter as low as 3 microns, and track the squeezed cell driven by a controlled pressure drop. This allowed us to simultaneously measure the variations of the RBC velocity as a function of the pressure gradient and of the local capillary diameter in a single experiment. At diameters below 5 microns, the membrane tension increases, thus effectively decreasing the cell's deformability, and the cell's velocity almost vanishes. In this regime, the velocity varies with the pressure head according to a characteristic power-law. We analyze our findings in terms of an elasto-hydrodynamic model for soft lubrication.

Multi-scale modeling of blood flows in extended coronary arteries

Simone Melchionna

Laboratory of Multiscale Modeling of Material, EPF Lausanne

The rheology of blood in large and medium-size vessels attracts much attention in view of understanding the physical basis of atherogenesis, the formation of atherosclerotic plaques in the endothelium. It is found that the occurrence of plaques strongly correlates with the regions of low shear stress in endothelial wall.
There are several physical factors contributing to modulating this behavior, such as the crowding of red blood cells near the endothelium and the complex interplay between hydrodynamics and suspended cells.
Red blood cells are by large the most important elements of bloods and their structural and dynamical response lay at the heart of multi-scale and multi-functional behavior.
In this talk, I will describe a strategy to simulate large-scale cardiovascular systems, with particular focus on coronary arteries.
Large-scale simulations up to tens of millions of red blood cells in fully blown coronary systems provide a wealth of novel information about the erratic motion of red blood cells, and their structural and dynamical heterogeneities, with crucial implications on the physiopathology of cardiovascular diseases.

In situ dynamics and fluidics of soft and biological matter

Thomas Pfohl

Departement Chemie, Universität Basel

The hierarchical self-organization of biological matter in cells, tissues, and organisms is one of the most fascinating phenomena in life science. Therefore, great efforts are devoted to elucidate the dynamics of the self-organization processes as well as to mimic these biological systems. As many biological processes consist of a series of transient steps in their reaction pathways that are undetectable in bulk measurements,

microfluidic-based experiments provide an opportunity to study the complexity of hierarchical dynamic and structural assembly and to generate models, which reproduce biological processes *in vitro.* Thus, detection in microfluidics can reveal transient steps far from equilibrium, even if they exist only briefly, revealing fundamental information about reaction mechanisms. By combining optical and spectroscopic microscopy, scanning small angle X-ray scattering and X-ray photon correlation spectroscopy with state of the art microfluidic technologies, we are able to characterize the self-assembly of chromatin-like materials, the bundling and network formation of filamentous proteins and their complex response on external stimuli and hydrodynamic flow on a mesoscopic as well as molecular scale.

Our studies on the behavior of soft biological objects under microflow conditions, such as cross-streamline migration and tumbling, may have apart from their fundamental relevance a great impact in biotechnical applications, such as analyzing and sorting individual molecules or organisms based on of their mechanical properties. In experiments with the unicellular parasite *Trypanosoma brucei* the impact of microscopic motility of on the overall swimming behavior of the parasite is analyzed. We observe that motility induced hydrodynamic drag forces help to protect trypanosomes against complement-mediated immune destruction in culture.

Theme 3: Colloids & Polymers 15.45 – 17.00

High sensitivity nanoscopy via interferometric scattering microscopy

M. Celebrano[a], P. Kukura[b], A. Renn[a], and V. Sandoghdar[a]

a) Nano-optics group, Laboratory for Physical Chemistry, ETH Zürich
b) Department of Chemistry, University of Oxford

High sensitivity detection of condensed matter is of fundamental importance in analytics and diagnostics at the nanoscale. The advent of single molecule fluorescence detection has already enabled a series of achievements that would not have been possible with ensemble techniques. However, all existing single-molecule methods require highly efficient fluorophores and, therefore, fail to access the great majority of species in our surrounding. As a result, recent works have reported high sensitivity detection based on the excitation of resonant pathways in matter (e.g. scattering, absorption, stimulated emission). Here we will report on the development interference scattering microscopy (iSCAT). This technique exploits the interference between the light scattered by a nano-object and that of the exciting beam to enhance the scattering (extinction) signal from the object itself. After an introduction on the basic concepts of iSCAT, I will focus on the experimental results obtained and the detection sensitivity achievable by this technique. In particular, I will concentrate on the detection and the spectroscopy of very small single plasmonic nanoparticles. Then, I will introduce specific experimental achievements in the fluorescence-free detection and tracking of dielectric nano-objects (i.e. single quantum dots, single viruses and proteins) and describe the progressive improvements of iSCAT sensitivity in the last years.

Geometry-induced trapping, levitation and assembly of nanometric objects in a fluid

M. Krishnan, N. Mojarad & V. Sandoghdar

Laboratory for Physical Chemistry, ETH Zürich

The ability to trap an object in space, whether a single atom or a macroscopic entity, is of primary importance in fields ranging from quantum optics to softcondensed matter physics, biophysics, and clinical medicine. Many sophisticatedmethodologies have been developed to counter the randomizing effect of Brownianmotion in solution, but stable trapping of nanometric objects remains verychallenging. For example, optical tweezing requires sufficiently polarizableobjects and is therefore unsuitable for manipulating small macromolecules. Anewer technique has succeeded in trapping single molecules, but this relies onactive feedback and does not offer nanoscopic confinement. Here we present afluidic trap concept that addresses these issues and provides trapping andlevitation of single charged nano-objects such as gold particles, polymer beads, andlipid vesicles for up to several hours. The principle is based on the spatialmodulation of the electrostatic potential created in a topographically-tailoredfluidic nanoslit. As a result, the trap requires no external intervention and is freeof constraints imposed by the objectâ€™s mass or dielectric function. The stiffness andstability of the trap can be tuned by varying the geometry of the system and theionic strength of the environment. The technique may be readily integrated withother manipulation mechanisms and holds great promise for contact-freeconfinement of single proteins and macromolecules, sorting and fractionation ofnano-objects, and the assembly of high-density arrays with applications rangingfrom biophysics to nanophotonics.

Diffusion dynamics of colloidal rods at liquid (solid) / liquid interfaces

Tahereh Mokhtari[a], Reinhard Sigel[a], Cristina Fernández-López[b], and Peter Lang[c]

a) Adolphe Merkle Institute, Université de Fribourg
b) Departamento de Química Física, Universidade de Vigo
c) Forschungszentrum, Institut für Festkörperforschung, Jülich

The dynamics of gold nanorods located within an oil-water interface and in the vicinity of a wall was studied using an evanescent wave dynamic light scattering (EWDLS) technique. The in-plane diffusion of the gold nanorods at the oil/water interface was found to be significantly slower than the bulk diffusion. In case of solid/liquid interface, the dynamics of the colloidal particles were comparable to the bulk diffusion. Ongoing theoretical work and computer simulations support this finding. Accordingly, a significant deviation from bulk dynamics should be observable only when the width of the rods is in the order of the evanescent wave penetration depth.

Prediction of the Attractive Double Layer Forces between Two Different Surfaces Using Charge Regulation Effects

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a) Department of Inorganic, Analytical, and Applied Chemistry, University of Geneva
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Direct force measurements between oppositely charged colloidal latex particles in aqueous electrolyte solutions were carried out with an atomic force microscope. Charge regulation effects describe quite well the observed attraction force profiles up to inter colloidal spacing of the order of Debye length. Moreover, when one of the surfaces is close to neutral, these effects even determine the sign of the double-layer force. The surface potentials and regulation parameters are determined from force data obtained in symmetric systems. We clearly show that charge regulation effects are essential in order to quantify double-layer forces in asymmetric systems with the Poisson Boltzmann theory.

Diffusion-limited reactions in crowded environments

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Diffusion-limited reactions are commonly found in biochemical processes such as enzyme catalysis, protein aggregation or complexation in cells. These processes are characterized by their reaction rate k which is the number of reactions per unit time. The simplest model of diffusion-limited encounter has been introduced by Smoluchowski under the hypothesis of infinite dilution and chemically isotropic spherical reactants. The Smoluchowski framework has been widely used to describe reactions occurring in vivo. However, biological environments are crowded: cell's cytoplasm, for instance, contains a

large number of proteins, nucleic acids, and other smaller molecules that can occupy up to 30-40% of the available volume. Thus, crowding effects are expected to impact considerably on the thermodynamics and kinetics of biological processes occurring within the cell.

We propose a first extension of the Smoluchowski framework that incorporates excluded volume effects, adapting Event Driven Brownian Dynamics to the particular configuration of an absorbing sink located at the center of a spherical bounding box and substrate spherical particles that diffuse around and get absorbed. The simulation scheme is designed to keep the density of substrate particles constant by reinsertion of the particles at the spherical boundary. For large absorbers, the reaction rate k obtained from the simulations can be accounted for through a pressure correction to the Smoluchowski rate. However, reducing the sink size (R_s), k is substantially depressed at intermediate packing and become even non-monotonic for $R_s << 1$. This saturation of the rate goes together a peculiar ordering of the diffusing particles in the vicinity of the absorbing center. A mean field analysis confirms that the R_s dependence of the rate is connected to an increasing competition between the diffusing particles when approaching a sink of smaller size. Finally, we show how the addition of an infinitesimal amount of non absorbing impurities can slow down dramatically the reaction.

Abstracts Posters

Theme 1: Materials & Assembly

T1-1: How membrane proteins act as gates for polymeric nanocontainers

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We tested the permeability of the membrane by encapsulation of a second enzyme inside the core of the vesicles, such as lactoperoxidase and studied its in situ activity, when its substrate was added from the environment of the vesicles.
These protein gates open large applications for controlled drug delivery by playing with the possible combinations of encapsulated molecules and sensitivity to environmental stimuli.
It is well known that one important function of biological membranes is to physically separate the intracellular constituents from the extracellular environment. Cell membranes are made of three major components: lipids, proteins and sugars.
We are trying to mimic biological membranes with artificial membranes made of block copolymers. Polymer membranes we are studying are made of the triblock copolymer PMOXA-PDMS-PMOXA (poly-2-methyl-2-oxazoline-poly(dimethylsiloxane)-poly-2-methyl-2-oxazoline). PMOXA-PDMS-PMOXA forms vesicular structures, and by reconstitution of membrane proteins, such as OmpF or FhuA we are able to create gates for molecules that do not passively diffuse across the polymer membrane. These channels allow the penetration of small molecules, up to 600 Da inside the aqueous cavity of the nanocontainers, without destroying the polymer membrane.

T1-2: Synthesis and Characterization of New Polyurea Elastomers by Sol-Gel Chemistry

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Polyurea chemistry is a relatively new synthetic process, similar to that used for the synthesis of polyurethanes. But unlike the polyurethanes, polyureas do not require a catalyst to accelerate the chemical reaction, due to the high nucleophilicity of amines. Polyurea chemistry is the result of a chemical reaction between a polyisocyanate and a polyamine to form urea groups that can interact by hydrogen bonding. Polyurea systems have better resistance to high pH, far superior thermal properties and an extremely higher melting point than the hybrids or polyurethane systems.
In this work, the synthesis of a new kind of polyurea elastomers by sol-gel chemistry is presented, where the reactivity of the amino-terminated polymers is reduced by the capping of the amines by reaction with ketones. This process allows controlling the gel time from seconds to minutes or hours. Upon drying the gel, an elastomeric films of defined thickness is obtained.
The resulting systems have been studied in terms of their mechanical properties and thermal stability, showing that the urea-crosslinking density and the architecture of the amino-terminated building block play an important role in their elastomeric behavior. This new type of elastomers combines the mechanical properties of physical and chemical networks.

Theme 2: BioPhysics

T2-1: Nanomechanical analysis of human cells exposed to the photo-oxidative stress

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Change in cell stiffness is a new characteristic of living cells exposed to various kind of stress. Using atomic force microscopy (AFM), we report the stiffness of live human cells, including fibroblasts, glioblastoma, bladder and melanoma cells exposed to the photo-oxidative stress generated in the presence of selected nano-structured photo-catalysts. The AFM experimental setup enabled us to generate the oxidative stress on living cells in situ, in a liquid-cell of the AFM scanner. We used low concentrations of nano-engineered particles (~10-6 M for fullerols and 0.05 - 0.5 ug/mL for nanoTiO2), as well as low intensities (~10 mW/cm2) of visible light (fullerols) and UV-A (nanoTiO2) illuminations. For cells exposed to the photo-oxidative stress, the AFM force spectroscopy revealed a marked drop in cell stiffness, which scaled with exposure to the deleterious action of ROS. These results point to the cytoskeleton reorganization occurring in mammalian cells at early stages of exposure to the nanoparticles-mediated photo-oxidative stress

T2-2: Probing the Glycocalyx with Photonic Force Microscopy

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The glycocalyx is a 3-D mesh of membrane-bound polysaccharides that envelop cell membranes externally. Depending on the type of cell, the glycocalyx can range from ~0.3 µm to ~4.5 µm in thickness. It is particularly thick in cells that are exposed to mechanical stress such endothelial and epithelial cells. In addition to its protective functions, the glycocalyx has also been shown to be important in mechanotransduction, hemostasis, cell-signaling, and blood cell-vessel wall interactions. It is also thought to play a role in a number of diseases such as diabetes, ischemia/reperfusion, and atherosclerosis. Due to its delicate composition, reliably imaging the glycocalyx and assessing its functionality has proved to be challenging.

With Photonic Force Microscopy (optical tweezers with back focal interferometric position tracking), we can probe the diffusion of sub-micron scale beads in 3D with micro-second time resolution and can also obtain 3D potential profiles encountered by the bead. With this technique, we are able to routinely probe the diffusive behavior of such a bead when in close proximity to a boundary surface, and have found very good agreement with theoretical predictions as given by Faxan's and Brenner's laws, for passive surfaces.

In this study, we have used PFM to probe, compare and contrast the glycocalyx influenced surface diffusion on cell surfaces of 2 types of human bladder epithelial cells: HCV29 (nonmalignant transitional epithelial cells of the ureter) and T24 (transitional cancer cells of the urine bladder). The glycocalyx is an important part of these cells as it protects them from the otherwise caustic environment that is the urinary bladder. We have also measured the interaction potentials of the surfaces in each of these cells.

T2-3: Statistical Analysis Study of AÎ² 42 Amyloid Fibril using Atomic Force Microscopy

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Amyloid plaques in the human brain are a well known manifestation of Alzheimer's disease. The major component of amyloid plagues is the fibrillar self-assembled amyloid Î²-protein (AÎ²). Many research groups proposed models for the amyloid fibril formation at each stage of fibrillogenesis, from soluble monomer to insoluble mature fibril. However there is still a lack of systematic statistical analysis of AÎ². In this work, we visualize the AÎ²1-42 fibrillogenesis at sequential incubation time and examine their structural behavior depending on the different substrate (HOPG/Mica) using atomic force microscopy. The determination of the flexibility and dimensions of the fibrils combined with the statistical polymer physics theory permit us to suggest a model for AÎ²1-42 fibril behaviors. The proposed model may be of general applicability to AÎ²1-42 fibril formation, and highlights the distinct flexibility of fibril depending on the maturation time.

T2-4: Manipulation of single supercoiled DNA molecules using Atomic Force Microscopy

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Although the hereditary information is encoded in the primary sequence of the genetic material, much of the regulatory information of DNA is hidden in its topology and geometry in the cell. DNA supercoiling and knotting are important for DNA packaging within all cells, however this impairs the well behavior of multiples cellular processes such as replication and transcription, which involve helical winding, strand separation, and movement along the DNA. The steric and topological problems associated with supercoiling can be studied with numerous different tools among which scanning probe microscopy is a very promising one because of its capability to image, on line and with a high resolution, biological samples in nearly physiological conditions.
In this work we studied the way enzymes and drugs interact with DNA molecules by using Atomic Force Microscopy (AFM). Especially we have centered our studies on human Type II DNA topoisomerases (Top2), which are essential and ubiquitous enzymes that perform important intracellular roles in chromosome condensation and segregation, and in regulating DNA supercoiling. We will present real time movies showing how the topo II relaxes DNA. In addition we will present AFM images showing the way DNA changes its topology after being exposed to intercalating agents. We hope that this new methodology will unveil some of the still the poorly known DNA-proteins and drugs interaction.

T2-5: Imaging and force spectroscopy on biological samples at LT and in UHV

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We performed imaging and force spectroscopy measurements on biological molecules (mainly dsDNA) in Ultra-High Vacuum (UHV) and at Low Temperature (LT). Experiments have been done by means of FM-AFM. We collected images of our samples varying the range of the tip-sample interaction forces and we tested several kinds of probes. We acquired Frequency shift vs. distance curves along the molecules, in order to derive local information about forces and interaction energy. We show preliminary results about: 1) the possibility of increasing the imaging resolution by setting properly the range of forces, 2) the observed heterogeneity of force curves along DNA.

T2-6: Photonic Force Microscopy

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Photonic Force Microscopy (PFM) is based on optical trapping and interferometric particle detection. It allows manipulation and multidimensional characterization of soft samples at the nanoscale, like living cells, polymer solutions and other kinds of fluids, viscous and elastic matter. Our user-friendly and compact prototype includes a powerful visualization and analysis software.

Theme 3: Colloids & Polymers

T3-1: Non-linear response of dipolar colloidal gels

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Dipolar colloids can be made to gel by forming a reversible but persistent network of chain-like structures. Using the model of charged soft dumbbells in molecular dynamics simulations, we find that, under the effect of an external electric field, this gel displays a highly non-linear electrical susceptibility. We show that this is due to the fact that a strong enough field make the system switch from a network to a structure of bundling chains. Such dramatic structural transformation upon applying external fields could allow the control of the mechanical and electrical response of these complex fluids, pointing to new applications of dipolar colloids as smart materials.

T3-2: A numerical study on 2 component colloidal gels

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A study of 2-components colloidal gels obtained using MD quench simulations is presented. The DNA-coated colloids are used as model. The two species are characterized by their relative concentrations c1 and c2 and a difference in the coupling explicit in the potential interaction, so that only colloids of the same specie feel a short-ranged attraction. The structures of the gel-like arrested systems are discussed and the conditions for obtaining a percolation of none, one or both the species are studied for systematic variations of c1, c2 and total packing fractions

T3-3: Effective forces in square well and square shoulder fluids

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We derive an expression of the effective force between a pair of macrospheres immersed in a sea of microspheres, in the case where the interaction between the two unlike species is assumed to be a square well or a square shoulder of given range and depth (or height). This formula extends a similar one developed in the case of hard core interactions only. Some qualitative features of such effective force and the resulting phase diagram are then analysed in the limit of no interaction between the small particles. Force profiles are then obtained by means of the integral equation theory combined with the superposition approximation and Monte Carlo simulations.

T3-4: Crystal structure of highly concentrated, ionic microgel suspensions studied by small angle X-ray scattering

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We present a small angle X-ray scattering study of the crystal structure formed by pH-sensitive poly(2-vinylpyridine) microgel particles with 5 wt% of cross-linker. We focus on highly swollen particles and explore concentrations ranging from below close packing to well above close packing, where the particles are forced to shrink or interpenetrate. The crystal structure found from poly- as well as mono-crystalline domains is random hexagonally close packed, as also observed in hard spheres.

T3-5: Depletion interactions in charged, aqueous colloid / polymer-mixtures

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Food and materials sciences have started to enormously profit from parallel developments in soft matter physics. Especially insight into model systems without specific interactions, like hard spheres, charged spheres, and ideal polymers, can easily be extrapolated to their industrial counterparts. In mixtures of these model components, steric exclusion of the polymer from an area around the colloid results in an attractive colloid-colloid interaction potential, the so-called depletion attraction. Theoretical models to predict depletion-induced phase behaviour apply both to organic model systems, as well as to mixtures of natural ingredients. In a next step towards reality's complexity we introduce electrostatic repulsions between the colloids, creating an interesting playing field to study phenomena like phase separation, transient gelation, and more exotic cluster formation. We systematically vary salt concentration and polymer size to cover a series of ratios between the ranges of attraction and repulsion.

T3-6: Influence of superplasticizers on cement suspensions

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Superplasticizers are chemical admixture usually applied to cement suspensions in order to improve their rheological properties. Despite many studies have been performed on them, their working mechanisms are not fully understood yet. Here we present a reading of the phenomenon which explains how the main effect of addition of superplasticizers to a suspension is to avoid precipitation of small particles on larger size colloids. These fine particles well-dispersed in the suspension fill the space between one big colloid and the other, avoiding their adhesion and allowing less friction among them. The theory is supported by experimental results obtained with atomic force microscopy (AFM), which shows different dispersion forces according to the charge of the particles, and with environmental scanning electron microscopy ESEM), which directly illustrates the decreasing of precipitation in presence of superplasticizers.

T3-7: Comparison of two cationic polymer architectures on the flocculation of negatively charged latex particles

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Our study point out the differences between the flocculation and the salt induced destabilization mechanisms. At optimal polymer dosage, the flocculation rate constants are significantly larger than the aggregation rate constant obtained with salt i.e. when charge screening has reached its optimal effect. The structure of the polymer is shown to play a significant role on the kinetics of flocculation, floc characteristics, and the range of concentration over which flocculation occurs. Optimal polymer dosage concentration is higher for the branched polymer which exhibits a greater flocculation rate constant, whereas the linear polymer is shown to have a larger concentration domain of use.

T3-8: Stimuli Responsive Dendronized Polymer: Large Mechanical Response Induced by Ionic Strength

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The capability of tuning mechanical properties of single polymer chain via external stimuli might represent a significant step toward their use as components in molecular machines.
In this work, mechanical properties of a new type of polymethacrylate-based dendronized polymers terminated with amine groups of different generations (PGn, n = 1-4) were studied with single molecule force spectroscopy. Our measurements show that the elastic properties of PG2, PG3 and PG4 change with the ionic strength. In particular, the chains soften strongly when the salt concentration is increased.

T3-9: Polyelectrolyte Behavior as a Function of pH, Salt Valency and Concentration. Monte Carlo Simulations.

F.Carnal and S. Stoll

F.-A. Forel Institute, Group of Environmental Physical Chemistry, University of Geneva

Acid/base and conformational properties of a weak polyelectrolyte chain surrounded by explicit ions are investigated using Monte Carlo simulations. Parameters such as the pH, salt valency or salt concentration play a key role in the titration process. The competition between attractive and repulsive, long-range and local electrostatic interactions leads to a heterogeneous distribution of charges and ions along the polyelectrolyte backbones. Focusing on trivalent salt, complex electrostatic interactions with chains occur leading to collapsed structures. The deprotonation process is promoted by the increase of salt concentration, which is also observed with monovalent salt particles.

T3-10: Low-Power Light Upconversion Schemes in Polymeric Matrices

Joseph Lott, Michelle Sing, **Yoan Simon** and Christoph Weder

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Light upconversion (UC) is a unique process whereby the wavelength of an incoming electromagnetic wave is shortened, effectively increasing the energy of the emitted light with respect to the incident one. One particularly interesting methodology to obtain blue-shifted light is upconversion by triplet-triplet annihilation (TTA-UC), as it allows one to work at low power densities (below 100 $mW.cm^{-2}$). Here, we present diverse strategies to incorporate upconverting dyads into rubbery polymer matrices. We investigated the influence of the glass transition temperature in the process and what it entails in terms of materials restrictions. We also introduce multicomponent systems as well as particle systems using poly-N-isopropylamide as a versatile thermoresponsive microgel. Finally, we look at potential strategies for photochemistry using TTA-UC and discuss the potential technological association of such systems.

T3-11: Reconstruction of smooth velocity and gradient fields from noisy scattered velocimetry data in a cross slot flow

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A substantial body of effort in recent years has been directed towards simulating the viscoelastic behavior of polymer melts and solutions in complex flow situations. Although several well known and widely used constitutive equations have been proposed, it is still quite difficult to accurately discriminate between the models and draw definite conclusions on their predictive capabilities.

In this regard, improvement can be achieved by measuring more accurately the flow fields. However, an inevitable feature of the experimental complex flow data obtained using particle tracking velocimetry and laser Doppler velocimetry is that it contains some undesired noise. Furthermore, evaluating constitutive equations to get the stress fields involves gradients of the velocity, which accentuates the challenge in obtaining significant results. Therefore, a proper judgment of the implications of the data needs applying appropriate regularization.

In this work, a generalized Tikhonov regularization combined with a finite element approximation is presented for reconstructing smooth velocity and velocity gradient fields from spatially scattered and noisy velocity data in a two-dimensional complex flow domain. Performances of diverse finite element continuity-regularization criterion combination are tested using synthetic velocity data in a cross-slot flow. This study shows that a careful choice of the finite element shape functions and the regularization matrix enables us to obtain an accurate, smooth, and continuous approximation of the velocity field and its gradient. Furthermore, optical fields are calculated by applying a differential constitutive equation directly to the reconstructed flow kinematics. The high quality of the reconstructed velocity gradient fields is shown to be an essential prerequisite for their reliable prediction. Overall, the method is expedient to implement and does not require boundary conditions.