

CeNS Workshop:

**EMERGING NANOSYSTEMS - FROM QUANTUM
MANIPULATION TO NANOBIMACHINES**



September 25th - 29th 2006

*Venice International University
San Servolo, Venice, Italy*



Workshop Schedule

Time	Monday 25-Sept-06	Tuesday 26-Sept-06	Wednesday 27-Sept-06	Thursday 28-Sept-06	Friday 29-Sept-06
9:30	<i>Opening</i>				
9:45	P.Hansma High speed atomic force microscopy and the molecular mechanics of bone fracture	H.E.Gaub Torturing molecules by AFM	T.W.Hänsch Towards a quantum laboratory on a chip	U.B.Wiesner Nanohybrids and nanobiohybrids	I.Frank Chemical reactions and dynamics in nanosystems: First-principles simulations
10:30	T.Basché Electronic excitation energy transfer between two single molecules	K.Hennessy Coupled excitons and photons in photonic nanocavities	D.Bensimon Single molecule studies of chromatin remodelling factors	J.Ebbecke Acoustic waves in semiconductor nanostructures	E.Yablonovitch The impedance-matching predicament: a hurdle in the race toward nano-electronics
11:15	<i>Coffee Break</i>				
11:45	F.Keilmann Spectroscopic near-field microscopy with infrared frequency-combs	T.Hugel Scrutinizing nanobiomachines with single-molecule fluorescence and force spectroscopy	P.Brouwer Current-induced magnetization dynamics in nano-scale magnets	S.Hoepfner Nanolithography and self-assembled monolayers – platforms to assemble functional structures	J.Feldmann Nanocrystals for photonic applications: towards synthetic light harvesting complexes
12:30	<i>Lunch</i>				
14:30	G.Abstreiter Manipulating and controlling semiconductor quantum dots	E.Gratton Particle tracking in 3D: chromatin structure and dynamics	<i>Informal Discussions</i>	J.E.Molloy Single molecule mechanical and optical studies: <i>in vitro</i> and inside live mammalian cells	<i>Departure</i>
15:15	E.Winfrey Algorithmic self-assembly of DNA	H.G.Craighead Nanostructures for biomolecular analysis		T.W.Ebbesen Surface plasmon photonics	
16:00	<i>Coffee Break</i>			<i>Coffee Break</i>	
16:30	W.M.Shih DNA-based molecular containers	T.Franosch Phase separation in driven non-equilibrium transport		V.Sandoghdar On the coupling of a single emitter to photons	
17:15	D.C.Lamb Single molecule investigations: from protein dynamics to virus entry	C.F.Schmidt Eg5: a four-headed mitotic kinesin motor		A.R.Bausch Physics of complex actin networks: from molecules to networks	
18:00	<i>Welcome</i>	<i>Dinner</i>		<i>Dinner</i>	
19:00	<i>Reception</i>	Poster Session I		Poster Session II	

Invited Speakers

ABSTRACTS

Manipulating and Controlling Semiconductor Quantum Dots

Gerhard Abstreiter

Walter Schottky Institut and Physik-Department Technische Universität München, D-85748 Garching, Germany

I will discuss the well controllable electro-optical properties of self assembled InGaAs quantum dots embedded in special Schottky diode structures. This includes the charge and spin storage, the coherent control of a single quantum dot photodiode as well as the controlled coupling of vertically stacked dots. The coherent control of charges, spins and photons are first steps towards semiconductor based quantum information technology.

This work is supported financially by German Research Association via collaborative research center 631 and by German Federal Ministry of Education and Research via NanoQuit.

The talk is based on the following recent publications:

- [1] M. Kroutvar et al., Nature 432, 81 (2004).
- [2] A. Zrenner et al., Nature 418, 612 (2002).
- [3] H. J. Krenner et al., New Journal of Physics 7, 184 (2005).
- [4] H. J. Krenner et al., Phys. Rev. Lett 94, 057402 (2005).

Electronic Excitation Energy Transfer Between Two Single Molecules

Thomas Basché

Institut für Physikalische Chemie, Johannes Gutenberg-Universität Mainz, Germany

Electronic Excitation Energy Transfer plays a key role in various molecular assemblies. The functioning of light-harvesting complexes and light-emitting devices composed of conjugated polymers crucially depends on the efficiency of this process. As has been shown in recent years, single molecule spectroscopy of molecular aggregates allows new insights into the fundamentals of electronic excitation energy transfer. Important aspects relate to the transfer mechanism(s) and the proper treatment of static disorder typically encountered in a solid state environment. Considering the case of Förster type energy transfer, an ensemble averaged spectral overlap J or Förster radius R_0 may not be a meaningful measure anymore for analyzing energy transfer at the single molecule level.

We have studied simple molecular dimers consisting of a donor (PDI) and an acceptor dye (TDI) separated by rigid oligophenylene spacers. Here, we present results of a donor-acceptor couple where PDI and TDI are linked by a p-terphenyl spacer. Due to the large spectral overlap, favorable dipole orientation and short interchromophore distance, energy is transferred quantitatively from the donor to the acceptor in this dyad.

By combining frequency selective single molecule spectroscopy and confocal fluorescence microscopy at 1.4 K, we have been able to deduce the energy transfer rates for single couples from line width measurements of the donor excitation spectra. Our approach allows for accurately taking into account static disorder in the donor and acceptor electronic transitions and to calculate the spectral overlap for each couple. By plotting the experimentally determined transfer rates against the spectral overlap, we can distinguish and quantify Förster and non-Förster type contributions to the energy transfer. The donor-acceptor couple studied to some extent mimics electronic coupling in conjugated polymers, which are viewed as a collection of chromophoric units separated by chemical defects or less conjugated segments. Consequently, energy transfer seems to occur as hopping of localized excitations the mechanism of which depends on interchromophoric distances and the nature of the “bridge”.

Physics of Complex Actin Networks: From Molecules to Networks

Andreas R. Bausch

Biophysik (E22), TU München James-Franck-Strasse D-85747 Garching, Germany

The cytoskeleton, a dynamic network of semiflexible actin filaments and associated regulatory proteins, is responsible for the extraordinary viscoelastic properties of cells. In order to understand the underlying physical principles it is important to determine *in vitro* the mechanical properties of entangled, cross linked and active actin networks on a microscopic scale. Besides the specific interactions in cells, unspecific physical interactions also drastically change the viscoelastic response of the cytoskeletal networks. Here we address the important aspects of geometrical confinement and relate the molecular structure of genetically designed crosslinker molecules to the structural and rheological properties of actin bundles and networks.

Single Molecule Studies of Chromatine Remodelling Factors

David Bensimon

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Chromosomes are DNA strings wrapped about histones “beads” (a structure known as chromatine) and compacted in the nucleus. The global accessibility of the chromosomes and the genes they code for is controlled by a large family of proteins known as chromatine remodeling factors. Some of these proteins chemically modify the histones thus altering their compaction in the nucleus. Others use ATP to “re-model” the chromatine. We will present results of AFM observations and single molecule measurements of the interaction of these proteins with bare DNA, i.e. without the histones. A simple model for the action of these proteins on DNA will be presented that accounts for much of the data.

Current-Induced Magnetization Dynamics in Nanoscale Magnets

Piet Brouwer

Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, NY 14853-2501, USA

Ferromagnets serve as spin filters for an electrical current passing through the magnet: the spin of the electrons that are transmitted through a ferromagnet becomes partially polarized parallel or antiparallel to the direction of the magnetization whereas spin current perpendicular to the magnetization direction is absorbed. Spin filtering is the root cause of the “giant magnetoresistance” effect, the observation that the resistance of a ferromagnetic multilayer depends on the relative orientation of the magnetic moments of the ferromagnets. It also is the origin of the “spin transfer torque”, the phenomenon that a spin polarized current impinging on a ferromagnet affects its magnetization direction. When combined, these two effects allow for all-electrical manipulation and detection of the magnetization state of nanoscale magnets.

In this talk, I discuss the physical mechanism of the spin transfer torque, the application of the spin transfer torque to magnetization reversal in ferromagnet–normal-metal–ferromagnet trilayers, and the use of the spin transfer torque to generate dynamical magnetization modes in ferromagnets.

Nanostructures for Biomolecular Analysis

Harold G. Craighead

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Advanced lithographic fabrication approaches can be combined with nanoscale-materials growth to form a wide range of structures and devices for interfacing to and observing biological systems. For example, electron-beam lithography and photolithography can be used to pattern biologically active molecular monolayers on solid surfaces, and these structures can be used to probe the response of cell signalling pathways. The engineered structure permits more quantitative interpretation of biological responses. Nanostructures can also form a variety of probes to transduce biological activity at high spatial resolution even allowing the real-time observation of individual active enzymes. Approaches we have studied include the utilization optical nanostructures for molecular-scale observation of activity in biological systems. We have been using such probes for studying biomolecular activity in living cell membranes and in model lipid-bilayer systems. We have also employed nanoscale fluidic systems and nanomechanical devices to isolate, enumerate and identify individual biomolecules of interest in small volumes.

Acoustic Waves in Semiconductor Nanostructures

Jens Ebbecke

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Surface acoustic waves (SAW) are mechanical waves on the surface of crystals that are accompanied in the case of piezoelectric crystals by an electric field. An electronic system in a semiconductor nanostructure mainly interacts with a SAW via this electric field. But the interaction strength is low while the electron density is high ("Fermi sea") because the electric field is screened. It can be regarded as an analogy to the tsunami (see Fig.1). As long as the sea has a large depth the amplitude of the tsunami is small. Only close to the coast this wave becomes dangerous.

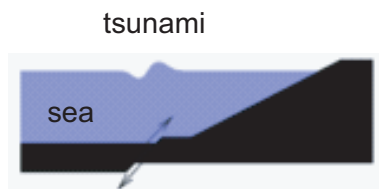


Fig.1 Schematic sketch of a tsunami.

Likewise if the electron density is reduced in a semiconductor nanostructure the electric field has a pronounced influence on the electronic properties. After an introduction of surface acoustic waves and a description of the interaction mechanism with semiconductor nanostructures examples of SAW mediated effects in two-dimensional electron gases, quantum wires and quantum dot will be presented.

Surface Plasmon Photonics

Thomas W. Ebbesen

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Materials structured on the nanometer scale can lead to improved and sometimes surprising properties. Metals are no exception to this rule. Metal particles for instance display colors which vary with their size. The color results from the coupling of light with the free electrons of the metal particle to form surface plasmons. On a broader level, with modern nanofabrication techniques it is possible to tailor the structure of metals and thereby to control the properties of surface plasmons opening many new possibilities from sensors to optical circuits [1].

The potential of surface plasmon photonics will be illustrated in some detail by our own work on the extraordinary optical properties acquired by metal films if they are simply structured with one or more apertures and periodic corrugations [2-8]. The different possibilities and challenges to create complete plasmon based photonic circuits will also be discussed [1,9,10]. Finally, the practical implications from chemistry to opto-electronics will be presented.

[1] Barnes, Dereux, and Ebbesen, *Nature* 424, 824 (2003). [2] Ebbesen et al., *Nature* 391, 667 (1998). [3] Martin-Moreno et al., *Phys. Rev. Lett.* 86, 1114 (2001). [4] Lezec et al., *Science* 197, 820 (2002). [5] Martin-Moreno et al., *Phys. Rev. Lett.* 90, 167401 (2003). [6] Garcia-Vidal et al., *Phys. Rev. Lett.* 90, 213901 (2003). [7] Barnes et al., *Phys. Rev. Lett.* 92, 107401 (2004). [8] Degiron et al., *Opt. Comm.* 239, 61 (2004). [9] Bozhevolnyi et al., *Nature* 440, 508 (2006). [10] Dintinger et al., *Adv. Mat.* 18, 1645 (2006).

Nanocrystals for Photonic Applications: Towards Synthetic Light Harvesting Complexes

Jochen Feldmann

Ludwig-Maximilians-Universität Munich

Chemically functionalized nanocrystals provide the unique possibility to build up hybrid super-structures with specific optical properties. The construction of controllable hybrid light harvesting complexes is one prominent example. In this talk some important steps towards their realization such as cascaded energy transfer [1,2] and electrical control of Förster energy transfer [3,4] will be discussed.

[1] T. Franzl et al., *Nano Letters* 4, 1599 (2004).
[2] T. A. Klar et al., *Adv. Mat.* 17, 769 (2005).
[3] J. Müller et al., *Nano Letters* 5, 2044 (2005).
[4] K. Becker et al., *Nature Materials* (in press).

Chemical Reactions and Dynamics in Nanosystems: First-Principles Simulations

Irmgard Frank

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Using First-Principles Molecular Dynamics in the Car-Parrinello scheme we are investigating the dynamics of various nanosystems in their ground and excited states. The approach allows to follow the nuclear motion in the quantummechanically computed electronic potential at finite temperature at the picosecond timescale. Besides dynamical phenomena such as the localization of chromophores in conjugated polymers, it is possible to simulate chemical reactions induced by mechanical load or by photoactivation.

- [1] D. Aktah and I. Frank, *J. Am. Chem. Soc.*, 124, 3402 (2002).
- [2] U. F. Röhrig, L. Guidoni, A. Laio, I. Frank, and U. Röthlisberger, *J. Am. Chem. Soc.*, 126, 15328 (2004).
- [3] C. Nonnenberg, C. Bräuchle, and I. Frank, *J. Chem. Phys.*, 122, 014311 (2005).
- [4] S. Grimm, C. Bräuchle, and I. Frank, *ChemPhysChem*, 6, 1943 (2005).
- [5] E. M. Lupton, C. Nonnenberg, I. Frank, F. Achenbach, J. Weis, and C. Bräuchle, *Chem. Phys. Lett.*, 414, 132 (2005).
- [6] C. Nonnenberg, H. Gaub, and I. Frank, *ChemPhysChem*, 7, 1455 (2006).
- [7] E. M. Lupton, F. Achenbach, J. Weis, C. Bräuchle, and I. Frank, *J. Phys. Chem. B*, in press.

Phase Separation in Driven Non-Equilibrium Transport

Thomas Franosch

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D-80333 Munich, Germany*

I will discuss aspects of collective behavior that emerge, e.g., in intracellular transport mediated by molecular motors like kinesin moving along cytoskeletal filaments. It has been recognized that such driven systems far from equilibrium may exhibit phase transitions sharing many features of their equilibrium counterparts. In particular, the totally asymmetric exclusion process (TASEP) has become the paradigm for boundary-induced non-equilibrium phase transitions. By weakly coupling TASEP to Langmuir kinetics unexpected phenomena such as phase separation and domain wall localization emerge. In a mesoscopic limit boundary and bulk dynamics become equally important, giving rise to rich phase diagrams. In the context of classical driven spin transport, the stationary state may exhibit a spin polarization phenomenon.

Torturing Molecules by AFM

Hermann E. Gaub

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The rapid development of novel experimental tools allowing the precise application and measurement of minute forces has opened exciting new perspectives in material- and life sciences. Particularly scanned probe techniques, which combine unparalleled spatial resolution with superb sensitivity, have allowed the development of a fundamental understanding of bio-molecular interactions and their nano-mechanical properties. Mechanical experiments with single molecules have become possible, and for the first time, intramolecular and intermolecular forces could be studied directly at the molecular level. Molecular recognition processes between many different ligand-receptor systems were meanwhile investigated and these studies have provided detailed insight in the underlying fundamental mechanisms. Even adhesion between live cells was resolved at the single molecule level. The option to investigate mechanical properties of biomolecules, such as polysaccharides, proteins and nucleic acids, attracted the interest of researchers, and in turn stimulated new developments in instrumentation. Proteins were unfolded at the resolution of individual helices with the precision of single amino acids. With the accessible force window, the whole range from entropic forces at several piconewtons (pN) to the rupture of covalent bonds at several nanonewtons (nN) can now be investigated. As single-molecule force spectroscopy continues to be a rapidly evolving field and now emerges as a widely used tool for the structural and functional investigation of bio-molecules in their native environment, this lecture will highlight the advances of the recent years.

Particle Tracking in 3D: Chromatin Structure and Dynamics

Enrico Gratton

University of California at Irvine, Irvine

The utilization of 2-photon microscopy in the field of Cell Biology is of increasing importance because it allows imaging of living cells, including those systems where UV imaging is not possible due to photobleaching or photodamage limitations. Using 2-photon excitation, the tracking method described in this work is suitable for studying the dynamics of single particles in cells or other systems sensitive to UV light.

We propose a novel approach based on the use of a scanner to produce an effective “intensity trap”. As the particle moves in this trap (note that there is no force applied on the particle at the power level we are using for particle detection), the detection system continuously calculates the position of the particle in the trap. As the position of the particle is calculated with respect to the trap, the scanner position is moved to minimize the “modulation” of the light intensity in the trap. In practice, we set the scanner to perform an orbit around the particle in about 1 millisecond. The sampling rate is chosen such that many points (32 or 64) are acquired during the orbit. An FFT (Fast Fourier Transform) is performed on the points acquired during one orbit or after a series of orbits, depending on the signal intensity. The DC, AC and phase of the first harmonic of the FFT are calculated. The value of the modulation (ratio AC/DC) varies monotonically as the distance of the particle from the center of the orbit is increased so that for every value of the modulation we can estimate the value of the distance of the particle from the center of the orbit. The phase of the first harmonic gives the angular position of the particle with respect to the scanner zero phase which is known relative to the lab coordinates. Of course, if the particle is too far from the trap, the DC value is very small and the modulation (although it remains high) loses its meaning. The algorithm responsible for the movement of the scanner senses the DC value, if it is below a selected threshold, then no movement is performed, but the radius of the orbit is slightly increased (or decreased) until some DC signal is detected. After detecting the DC signal, we know the position of the object causing the signal and the scanner is immediately centered at this position. This simple algorithm allows us to initiate tracking of the first bright spot encountered in the vicinity of the starting scanner position. The effective bandwidth of the tracking system depends on the maximum frequency for sinusoidal oscillation of the scanner, which is about 5 kHz for our galvano-scanner and on the number of photons needed for detecting the particle against the noise. In our algorithm, more orbits can be averaged if the signal is very weak thus effectively decreasing the bandwidth of the tracking apparatus. For example, a single molecule such as fluorescein or rhodamine can provide enough photons in one millisecond to allow tracking. Of course, there are other important considerations. First, if the motion of the particle is too fast such that after one orbit the particle moves too far from the new position calculated based on the previous orbit, tracking is lost since the feedback mechanism is too slow. Therefore, single molecules, which in water would move across the PSF in about 0.1 ms, cannot be tracked. We need at least a macromolecule the size of a large protein (100kD) or relatively high viscosity to increase the time a fluorescent particle can be observed in the PSF. The second consideration, perhaps the most important, is that the particle should not bleach during the length of the tracking. This is not a problem for particles made of many fluorophores, such as polystyrene fluorescent beads, which are also relatively large. Surprisingly, in all biological systems we investigated, photobleaching did not occur.

Towards a Quantum Laboratory on a Chip

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Microfabricated magnetic traps, waveguides, and other elements for the manipulation of ultracold atoms can be combined to form a quantum laboratory on a chip. Devices such as miniaturized atom lasers, atom interferometers, and atomic clocks have been implemented in this way. Atom chips are also offering intriguing perspectives for quantum simulations and quantum information processing. Recent experiments at the University of Munich (LMU) will be reviewed.

High Speed Atomic Force Microscopy and the Molecular Mechanics of Bone Fracture

Paul Hansma

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It is now possible to operate Atomic Force Microscopes (AFMs) at speeds of up to 6000 lines per second over scan ranges exceeding 10 microns. For a 100×100 pixel image this gives frame rates of 60 frames/second: faster than video rate. This has required small cantilevers, new scanners, new high voltage amplifiers and new control algorithms. This talk will focus on the new scanners and control algorithms. A new high speed AFM as well as more conventional AFMs and SEMs have given new information about the molecular mechanisms and mechanics that are involved in bone fracture. High resolution images suggest that the fundamental mechanism for bone fracture is separation or relative motion of mineralized collagen fibrils. This separation or relative motion is resisted by “glue”, which AFM pulling reveals to use the sacrificial bond-hidden length mechanism that has previously been shown to operate in many biological systems such as in abalone shells and spider silk. Osteopontin, a glycoprotein which tends to be highly phosphorylated in bone, appears to be one of the components of the glue in bone. The phosphate functional groups on the osteopontin appear to be involved in the sacrificial bonds.

Coupled Excitons and Photons in Photonic Nanocavities

Kevin Hennessy, Antonio Badolato, Martin Winger, Ataç Imamoğlu, and Evelyn Hu

U. of California, Santa Barbara CA 93106 & ETH-Zürich, 8043 Zürich

We have developed a fabrication technique that allows us to position single InAs quantum dots (QDs) at precise locations in photonic crystal nanocavities. With the QD position in the cavity known, we have studied the interaction between a discrete QD exciton state and the vacuum photon state of the cavity. By bringing the two states into spectral resonance, we observe clearly the strong coupling regime between the exciton and photon, resulting in the creation of two coupled polariton states. Such states presage many fundamental applications in quantum information processing.

While the formation of polaritons agrees well with cavity-QED predictions in our devices, we observe evidence of exciton/cavity coupling of a different nature that is not explained by current models. These unexpected findings, which strongly challenge the notion of QDs as “artificial atoms”, will be discussed.

Nanolithography and Self-Assembled Monolayers – Platforms to Assemble Functional Structures

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Laboratory of Macromolecular Chemistry and Nanoscience, Den Dolech 2, 5600 MB Eindhoven,
The Netherlands

The patterning of surfaces by means of scanning probe microscopy techniques has attracted a lot of interest during the last years [1]. The small structure sizes that are obtainable by means of the nanoscopic probe tips allows the manipulation of surfaces with nanometer precision. This is doubtlessly regarded as one of the crucial tasks that has to be addressed for nanofabrication approaches.

We demonstrate a chemical bottom-up fabrication process that utilizes the electro-oxidative functionalization of self-assembly *n*-octadecyl trichlorosilane (OTS) monolayers by means of an electrically conductive scanning force microscope tip [2]. The surface terminal $-CH_3$ functions are electrochemically converted into reactive $-COOH$ groups when a negative tip bias voltage is applied to the tip.

These surface areas can be chemically addressed and serve as a template for subsequently performed chemical derivatization routines to hierarchically assemble functional nanostructures. Examples include e.g. the generation of nanoparticles on the surface template or the binding of nanomaterials such as magnetite particles and nanotubes. Moreover are such templates also capable to mediate/initiate chemical reactions. Examples for this approach include the site-selective growth of polymer brushes on chemically active surface templates, or “click” reactions on functionalized substrates.

Different modification routines will be demonstrated which have the potential to be implemented within hierarchical, step-by-step fabrication approaches towards the planned fabrication of nanodimensional functional devices.

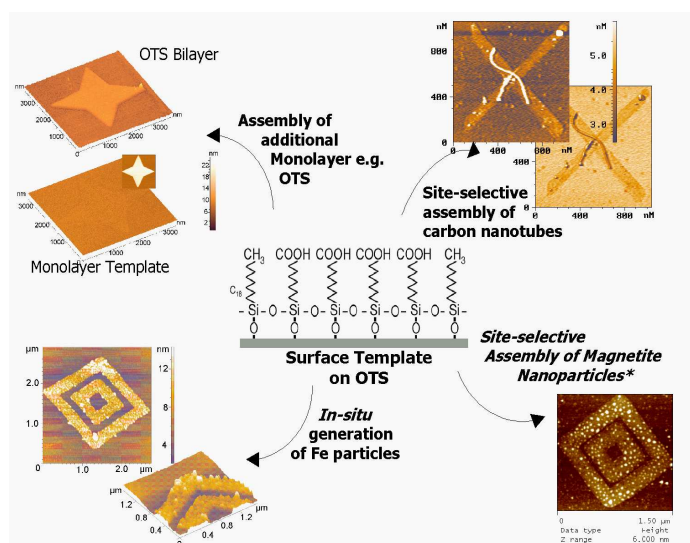


Fig.1 Chemically active surface templates generated by Electro-oxidative Nanolithography can be used for different modification schemes. Different examples are depicted to emphasize the potential of the method in terms of nanofabrication.

[1] D. Wouters, U. S. Schubert, *Angew. Chem. Int. Ed.* 43, 2480 (2004).

[2] R. Maoz, E. Frydman, S. R. Cohen, and J. Sagiv, *Adv. Mater.* 12, 725 (2000).

[3] S. Hoepfener, R. Maoz, S. R. Cohen, L. F. Chi, H. Fuchs, and J. Sagiv, *Adv. Mater.* 14, 1036 (2002).

[4] S. Hoepfener, U. S. Schubert, *Small* 1, 628 (2005).

Scrutinizing Nanobiomachines with Single-Molecule Fluorescence and Force Spectroscopy

Thorsten Hugel

Biophysics (E22) and Institute for Medical Engineering (IMETUM), TU Munich, Garching, Germany

A thorough understanding of nature's fascinating molecular machines will not only help to develop better drugs, but should also guide the construction of man-made nanosystems, especially if they have to function in physiological conditions. We report on our investigations on two molecular machines, the portal import motor of the bacteriophage Phi29 and the molecular chaperone HSP90.

The bacteriophage Phi29 generates large forces to compact its dsDNA genome into a small protein capsid by means of a portal motor complex [1]. Several mechanical models for the generation of these high forces by the motor complex predict coupling of DNA translocation to rotation of the head-tail connector dodecamer. Putative connector rotation is investigated here by combining the methods of single-molecule force spectroscopy with single-molecule fluorescence polarization [2].

The molecular chaperone Hsp90 is involved in folding, activation and stabilization of various substrate proteins [3]. It consists of two monomers that undergo major conformational changes during ATP Hydrolysis. We investigate these conformational changes as well as the binding of fluorescence labelled ATP and substrate proteins by fluorescence resonance energy transfer (FRET).

[1] Y. R. Chemla et al., *Cell* 122, 683 (2005).

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[3] H. Wegele, L. Müller, J. Bucher, *Rev. Physiol. Biochem. Pharmacol.* 151, 1 (2004).

Spectroscopic Near-Field Microscopy with Infrared Frequency-Combs

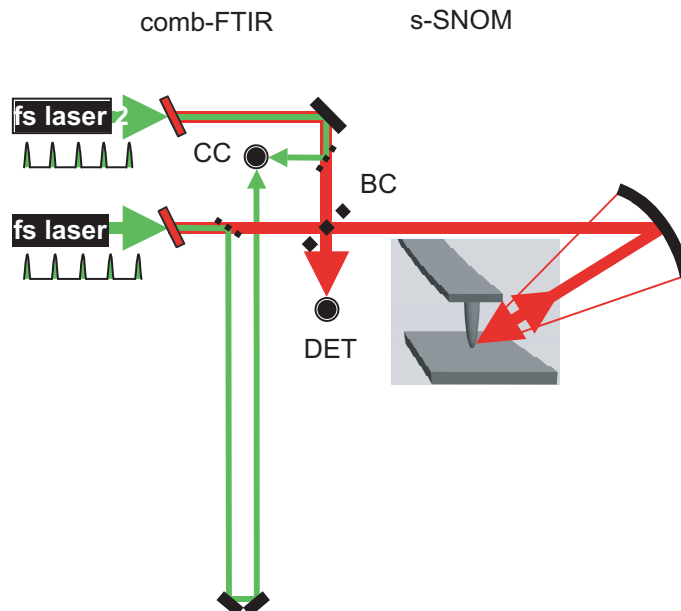
Fritz Keilmann, Markus Brehm, Albert Schliesser

Max-Planck-Institut für Biochemie & Center for NanoScience, Martinsried (München), Germany

We advance scattering scanning optical near-field microscopy (s-SNOM) that resolves 10 nm features independently of the wavelength into broadband-spectroscopic operation. We concentrate on the mid-infrared region with wavelengths 5-15 μm where materials can be recognized by “fingerprint” molecular absorption and conduction phenomena.

The “aperture-less” s-SNOM uses a standard, metallized AFM tip that serves the additional role of an infrared antenna, generating a highly confined near-field spot at the tip apex. The near-field interaction with the sample influences the back-scattered light such that its amplitude and phase enables the determination of the local complex-dielectric value [1]. In the past we have employed step-tunable gas lasers to collect s-SNOM images at varied wavelength settings. Local spectra of nanosystems could then be computed off-line, showing Drude response of buried charge carriers [2], “reststrahlen” resonance of lattice vibration [3], and molecular vibrational resonance in polymers [4]. Mapping the amide I vibrational resonance of a single virus revealed a highly enhanced sensitivity over plane-wave absorption spectroscopy [5]. A drawback with gas lasers is the lengthy procedure of repeated imaging, prone to errors from sample drift and from tip erosion.

We have recently introduced coherent frequency-comb infrared spectroscopy that uses 70 000 evenly-spaced mid-infrared frequencies spanning 9-12 μm [6]. Two such sources are used for self-scanning interferometry. A rapid sequence of up to 1000 mid-infrared spectra/s has been recorded without moving parts [7]. Here we report successfully combining comb-FTIR with s-SNOM as sketched in the figure. Our future aim is to achieve spectroscopic s-SNOM for an even broader mid-infrared region, for general chemical and structural recognition of nanoscale objects.



- [1] R. Hillenbrand et al., Phys. Rev. Lett. 85, 3029 (2000). [2] B. Knoll et al., Appl. Phys. Lett. 77, 3980 (2000).
[3] R. Hillenbrand et al., Nature 418, 159 (2002). [4] T. Taubner et al., Appl. Phys. Lett. 85, 5064 (2004).
[5] M. Brehm et al., NanoLett. 6, 1307 (2006). [6] F. Keilmann et al., Opt. Lett. 29, 1542 (2004).
[7] A. Schliesser et al., Opt. Express 13, 9029 (2005).

Single Molecule Investigations: From Protein Dynamics to Virus Entry

Don C. Lamb

LMU München, Department Chemie und Biochemie

Single molecule techniques have developed over the last decade from the detection of single molecules to an important method in Chemistry, Biochemistry, and Biophysics. Here, we present two areas where single molecule techniques have brought advances to biophysical understanding.

Using single-pair fluorescence resonance energy transfer, we have investigated the dynamics of the TATA-Box -Binding Protein (TBP) during binding to DNA and how the dynamics are influenced by interactions with an inhibitory protein, negative cofactor 2 (NC2). After the initial binding steps, TBP is bound with a well defined position on the DNA. Upon addition of NC2, the TBP-NC2 complex becomes mobile on the DNA, shifting rapidly between different positions and/or conformations. This dynamics brings an entirely new perspective to the mechanistic understanding of gene regulation which was previously thought to occur via steric hindrances by NC2.

Single molecule techniques can also be applied to the study the entry pathway of individual viruses and non-viral vectors in living cells. Non-viral vectors provide a flexible system for designing and constructing gene-transfer vectors. The uptake of these gene-transfer vectors into the cell, their transport and their interactions with the cytoskeleton have been characterized in detail. Upon mitosis, their transport along astral microtubules towards the poles of the spindle apparatus during mitosis has been observed as well as the successful infection of the cells after division.

Single molecule mechanical and optical studies: *in vitro* and inside live mammalian cells

Justin E. Molloy

MRC National Institute for Medical Research, London, UK

We have used an optical tweezers based transducer to study the mechanism of force generation by actomyosin I and have used total internal reflection fluorescence microscopy (TIRFM) to investigate the targeting and trafficking of GFP-tagged myosin X within live mammalian cells.

Our optical tweezers experiments made using non-muscle, myosin I shows that the myosin power-stroke is generated in two discrete phases [1,2]. This result is interesting from a mechanistic standpoint as it shows myosin can generate part of its movement with a delay after binding to actin (the so-called “rocking cross-bridge” hypothesis). Furthermore, analysis of the actomyosin bound lifetime distribution shows that there are two detachment pathways from the tightly-bound actomyosin state. There is a fast, ATP independent, detachment pathway and a much slower, ATP dependent pathway (“weak” and “strong” interactions). Both of these discoveries might contribute to the physiological role of Myo1c in sensory adaptation in the inner ear [3]: “rocking” between actomyosin bound states might account for fast adaptation and slower, long-range slipping adaptation might be due to “weak” interactions of myo1c with actin.

Myosin-X is an actin-based molecular motor that is recruited to the tips of filopodia in live, cultured, cells [4]. Three pleckstrin homology domains (PH123) in the tail region of this myosin are thought to be responsible for its targeting to plasma membrane. We have used TIRFM to measure the binding kinetics of single, GFP-tagged, PH123 molecules at the plasma membrane of live mammalian cells and found a dissociation rate of 0.05 s^{-1} (i.e. they remain bound for about 20 sec [5]). We have also measured the lateral diffusion coefficient of GFP-tagged PH123 and full-length myosin-X by tracking individual fluorophores moving on the inner leaflet of the plasma membrane. PH123 molecules showed significantly faster lateral diffusion ($0.2 \mu\text{m}^2\text{s}^{-1}$) than myosin-X ($0.08 \mu\text{m}^2\text{s}^{-1}$). However, mild treatment with the actin filament disrupting drug, latrunculin-B, increased mobility of intact myosin X to $0.12 \mu\text{m}^2\text{s}^{-1}$ but had no effect on PH123 mobility. Myosin X is targeted to phosphoinositol-phospholipids (mainly PI(3,4,5)P3) by its PH domains but its mobility is additionally controlled by motor domain interactions with the actin cytoskeleton.

- [1] Veigel et al., *Nature*, 398, 530 (1999). [2] Batters et al., *EMBO J.*, 23, 1433 (2004).
[3] Holt et al., *Cell*, 108, 371 (2002). [4] Berg & Cheney, *Nat. Cell. Biol.*, 4, 246 (2002).
[5] Mashanov et al., *J. Biol. Chem.*, 279, 15274 (2004).

On the Coupling of a Single Emitter to Photons

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The interaction of atoms and molecules with light is often described in terms of an absorption cross section, which is often considered to be an intrinsic property of the emitter. We discuss two different experiments where we enhance the coupling between a single molecule and light. In the first case, a single gold nanoparticle acts as a nano-antenna to modify the excitation, radiation and dissipation processes of a single molecule [1]. The second experiment examines the coherent interaction of strongly confined light with a single molecule at cryogenic temperatures [2]. Our results show that a single molecule can leave a fingerprint of well beyond 10% on a laser beam.

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<http://arxiv.org/abs/quant-ph/0604177>.

Eg5: A Four-Headed Mitotic Kinesin Motor

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Cell division is organized by the meiotic or mitotic spindle, which is a highly complex nanomachine. Forces are generated both by microtubule polymerization/depolymerization and by motor proteins. We have investigated the tetrameric kinesin motor Eg 5 which is essential for spindle morphogenesis. We have used both single-molecule fluorescence assays and single molecule laser trapping assays. Eg 5 shows both diffusive and processive behavior and the switching between the two modes appears to be controlled by binding to two microtubules. The motor also appears to behave quite differently from kinesin 1 in terms of its load dependence. We hypothesize that observed load-dependent release at relatively low forces might be relevant as a force-sensing device in the mitotic spindle.

DNA-Based Molecular Containers

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Molecular containers with cavities on the 10-100 nm scale could have many applications, including drug delivery, capture and stabilization of transient multiprotein complexes, and provision of microenvironments for processes such as protein folding. Towards these ends, investigation of three architectural strategies for building DNA-based molecular containers, each offering different application-specific advantages, has been initiated. These strategies use either scaffolded or single-stranded DNA origami approaches. (i) Scaffolded thirty-helix bundle hexagonal cylinders, when enclosed with caps, would be expected to resist permeation into their cavities of molecules larger than one nanometer in diameter, due to the small sizes of their pores. Scaffolded cylinders with heights of ≈ 45 or 60 nm and diameters of ≈ 20 nm have been generated, as have scaffolded flat stamps with dimensions of $\approx 70 \times 50$ nm. The stamps potentially could serve as caps for the cylinders. (ii) Scaffolded wireframe deltahedra with six-helix bundle DNA-nanotube struts should have large strength-to-weight ratios, and may be adaptable for generating rigid containers with cavities on the 100-nm or larger scales. Tetrahedra in this design have been generated with struts that are ≈ 40 nm long. (iii) Single-stranded DNA wireframe octahedra and tetrahedra have the potential to be expressed and folded within cells. Octahedra and tetrahedra in this design have been generated with struts that are either 11 or 14 nm long. Analyses with agarose-gel electrophoresis and negative-stain electron microscopy indicate that, for each of the three target classes, the majority of molecules fold into monodispersed particles whose shapes resemble the target folds.

Nanohybrids and Nanobiohybrids

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Polymer based nanostructured organic-inorganic hybrid materials constitute a fascinating research area with tremendous scientific as well as technological promise. By choice of the appropriate synthetic polymers as well as inorganic species unprecedented morphology and functionality control down to the nanoscale is achieved. For example, we recently demonstrated lithium ion transport in nanoscopic (10 nm) domains of a bicontinuous cubic morphology (see Fig.1) [1]. In polymer-ceramic hybrids the interface is of key importance. The hydrophilic parts of the polymer can be completely integrated into the hybrid ceramic composite phase. Through this unique interface engineering properties of polymers can be uniquely married with those of solid-state materials. Once the formation mechanisms involving co-assembly of polymeric and nanoparticle (sol) species are understood, morphologies across the entire phase space of blocked amphiphilic macromolecules can be produced. A particularly fascinating bicontinuous cubic structure we discovered is referred to as the “Plumber’s Nightmare” and its inverse [2].

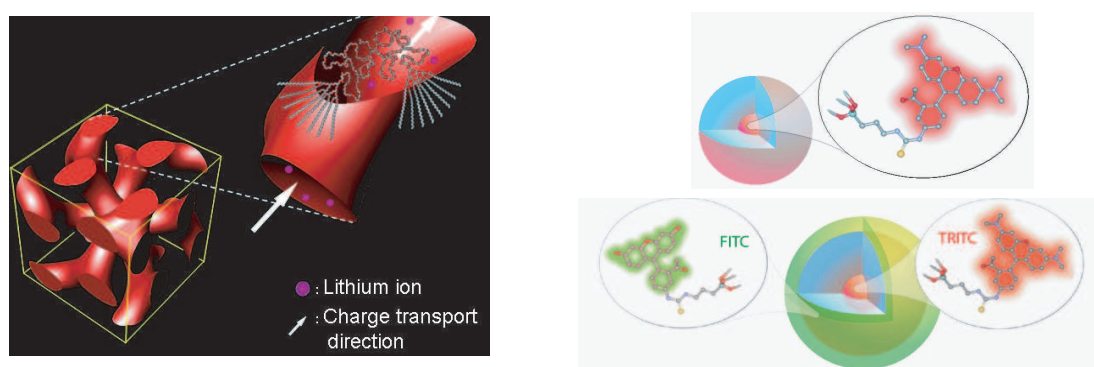


Fig.1 Charge transport in a nanostructured double gyroid [1]. **Fig.2** Cornell Dot Architectures.

Completely novel property profiles are accessible through polymer-inorganic hybrids. For example, the results in Fig.1 suggest an advanced molecular design concept for the next generation nanostructured materials in applications involving charge transport (e.g. ion conductors, photovoltaics, fuel cells). Mesoporous materials with crystalline, superparamagnetic iron oxide nanoparticles in the walls obtained through high temperature treatment of polymer-inorganic hybrids open a way towards separation media exhibiting a combination of size exclusion and separation based on magnetic interactions. The approach can be extended from linear to dendritic, and from AB to ABC-type architectures. On the inorganic side besides amorphous and crystalline oxide materials novel systems toward nanostructured high temperature SiCN and SiC can be introduced. Finally, the approach is successfully extended to thin films as well as to fluorescent hybrid silica nanoparticles (Cornell Dots) for applications in Nanobio-technology [2]. Fig.2 shows two architectures for Cornell dots as fluorescent labels as well as ratiometric single particle pH sensors for applications, e.g., in molecular-scale imaging and high throughput pharmaceutical screening.

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Algorithmic Self-Assembly of DNA

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I will review recent progress implementing algorithmic self-assembly processes using DNA tiles and DNA origami, focussing on issues of pattern formation, information copying, self-assembly error rates, including proofreading and designed nucleation barriers.

The Impedance-Matching Predicament: A Hurdle in the Race Toward Nano-Electronics

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In contemplating the headlong rush toward miniaturization represented by Moore's Law, it is tempting to think only of the progression toward molecular sized components.

There is a second aspect of Moore's Law, that is sometimes overlooked. Because of miniaturization, the energy efficiency of information processing steadily improves. We anticipate that the energy required to process a single bit of information will eventually become as tiny as 1 electron Volt per function, truly indeed a molecular sized energy. Inevitably most logic functions including storage, readout, and other logical manipulations will eventually be that efficient. However there is one information-processing-function that bucks this trend. That is communication. Our best projections of improvements in the electrical communication function shows that it will still require hundreds of thousand of electron Volts, just to move a bit of information a relatively short distance.

Why this energy per bit discrepancy for communications? It is caused by the difference in length scale between the wires and the nano-sized devices. Wires are long and that leads to a low impedance, devices are tiny and they have a high impedance. The challenge then is to find, discover, or invent a nano-scale impedance transformer, that can bring the energy efficiency of communications in line with the excellent efficiency of all the other bit functions.

One such impedance matching concept is optical communications itself. Optics might substitute for the longer wires known as global interconnects. In this respect it is important to be aware of the balance between short and long wires on the chip. Wire length properties are summarized by a single dimensionless exponent [1] in Rent's rule. Efficient long electrical interconnects would require lower signaling voltages, but that would slow down the individual wires, and would demand excessive parallelism of long wires to keep up the data rate. Thus the "efficient" electrical solution is not practical since it would require an excessive density of long wires, and an unrealistic Rent's rule exponent. This will create a power advantage for optical signaling on-chip.

[1] Rent's rule is explained in "The Feynman Lectures on Computation", in the last six pages of the text.

Posters



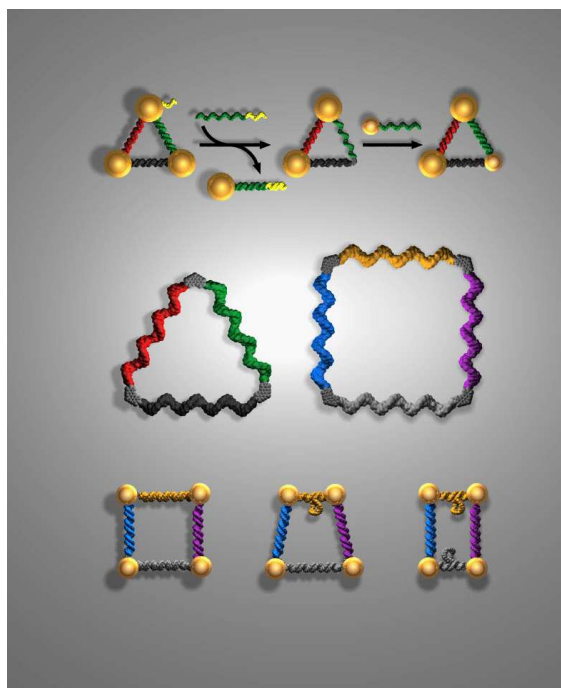
ABSTRACTS

DNA-Mediated Organization of Nanoparticles Into Discrete Assemblies: Control of Geometry, Modularity, Write/Erase and Structural Switching

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Gold nanoparticle assemblies have recently emerged as a promising class of materials with novel optical, electronic, catalytic and sensing applications. In order to carry out fundamental studies of these phenomena, a general method to organize particles into finite well-defined assemblies of pre-determined geometries needs to be developed. In here we discuss a method for the organization of gold nanoparticles into a well-defined two-dimensional *discrete* assembly, in which six gold particles are arranged into a hexagon. The approach involves labeling the individual nanoparticles with DNA containing building blocks that serve to dictate their location within the final constructs. A second generation system that differs fundamentally in that the template is fully functional and separate from the nanoparticles is also discussed in detail. We show how a fully cyclized single stranded DNA template, containing rigid organic molecules as the vertices, allows for the modular organization of gold nanoparticles with control over both geometry and modularity. We also show how the same templates are geometrically dynamic and how they possess a post-assembly write/erase function. Such a dynamic template provides a scaffold on which a large number of nanoparticle constructs is readily accessed, and allows for the ability to truly realize the potential of “single electronics” and “nanoscale optics”.



Electrical Control of Förster Energy Transfer

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Bringing together compounds of intrinsically different functionality such as inorganic nanostructures and organic molecules constitutes a particularly powerful route to creating novel functional devices with synergistic properties found in neither of the constituents. We introduce nanophotonic functional elements

combining two classes of materials, semiconductor nanocrystals and organic dyes, whose physical nature arises as a superposition of the properties of the individual components.

The large absorption cross section of the nanocrystals provides an efficient route to concentrating excitation energy and photopumping single dye molecules with much weaker absorption cross sections. The CdSe/CdS nanorods employed exhibit a large quantum confined Stark effect (QCSE) [1,2], which enables direct control of the spectral resonance between donor and acceptor required for Förster-type energy transfer in single nanorod-dye couples [3]. With this far-field manipulation by a remote electric field of a near-field phenomenon, the emission from single dye molecules can be controlled electrically [3]. Such a device constitutes a novel single molecule switch, resembling an optical field-effect transistor shuttling photons rather than electrons.

Interestingly, the efficiency of both synthetic light-harvesting systems and of natural complexes is generally limited by energetic disorder between different chromophores. This can lead to detrimental trapping of excitation energy in local energetic minima. As a result, a macroscopic description of energy transfer rates in inhomogeneously broadened ensembles is often misleading. Electrically tailoring the individual spectral overlaps of the optical transitions of donors and acceptors in energy transfer cascade structures may offer a route to a deeper understanding of the functioning of such light-harvesting complexes.

[1] J. Müller et al., Nano Lett. 5, 2044 (2005). [2] J. Müller et al., Phys. Rev. Lett. 93, 167402 (2004).

[3] K. Becker et al., Nature Materials (submitted).

Fabrication of Harmonic Generation and Two-Photon Fluorescence Microscope

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We fabricated a harmonic and two-photon fluorescence microscope. Due to the low scattering of infrared laser light in living cells, it is able to image samples at depths up to 500 μm . Moreover, the longer wavelengths used are less harmful to biological samples and in many cases, no labeling is necessary.

Second harmonic generation (SHG) has proven to be present in non-centrosymmetric medium and third harmonic generation (THG) occurs mainly at interfaces. Since both effects are complementary, it is interesting to use both of them with the addition of two-photon fluorescence (2PF).

We also present multiphoton images from lilies' pollen, fluorescent beads, rat tail tendon and 40 nm gold nanoparticles.

Optical Spin Initialization and Readout in a Single Self-Assembled Quantum Dot

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Single electron spins in self-assembled quantum dots are a promising candidate for realizing quantum bits in semiconductor devices. Compared to excitons, the spin of an electron in a quantum dot has a long lifetime [1,2], a crucial prerequisite for spin manipulation. Furthermore, self-assembled quantum dots can be charged with single electrons in a controlled way by applying a gate voltage. We probe the quantum dot ground state by high resolution laser spectroscopy of the resonant Rayleigh scattering [3]. This technique allows for direct probing of the quantum dot spin states, by controlling the polarization of the excitation light [3]. We show that the scattering signal is strongly dependent on the population of

the quantum dot spin state and that the quantum dot can be initialized in the spin up and the spin down state. This can be achieved either by thermal occupation of the energetically lower spin state [4], or by Raman mediated pumping of the electron spin into the upper spin state [5]. The latter offers a promising spin initialization method since it provides high fidelity (>90%) at low ($B < 1\text{T}$) magnetic field strengths. This opens up the possibilities of microwave induced spin flips at reasonable frequencies.

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Single Molecule Unzipping of Coiled Coils: Sequence Resolved Stability Profiles

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We use high resolution atomic force microscopy to mechanically unzip and rezip single coiled-coil proteins. This allows us to read off the complete stability profile of the protein turn by turn. We investigated three coiled coils with different length as well as a point mutation and find force fluctuations between 9 and 15 pN that can be directly related to the amino-acid sequences. An equilibrium model previously applied to DNA fully describes the mechanical unzipping process including free-energy contributions of the individual turns and seed formation energy. We want to use this technique to explore stability profiles of artificial as well as physiological relevant coiled coils. In special we want to compare the neck regions from fungal and animal kinesins, because this coiled coil motives are proposed to influence motor properties.

Convective Replication

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Thermal convection gives a twofold new approach to the origin of life. Only by applying a temperature gradient, we could achieve in the same convection chamber a PCR reaction driven by the temperature cycling of the convection flow and thermophoretic trapping of DNA. Combined it would elegantly realize the two physical problems of prebiotic evolution (replication and accumulation) in a primitive natural setting of heated pores.

The chain reaction of PCR relies on temperature cycling. Here, we demonstrate that a thermal convection state can drive DNA replication. We induce thermal convection with a temperature gradient in a millimeter-sized chamber. The laminar flow ensures a reliable temperature cycling. We amplify a single product 100,000-fold after 25 minutes. The replication is exponential, doubling every 55 s.

We measured for the first time the thermophoretic repulsion of DNA using fluorescence sensing of temperature in a thin, convection-free chamber. We measure a thermal diffusion constant $D_T = 0.4 \times 10^{-8} \text{ cm}^2/(\text{sK})$. In a thicker chamber, convection overcomes the repulsive force of thermophoresis near the chamber wall and accumulates DNA more than thousandfold towards the wall at the heating source. Both mechanisms use the negative-entropy source of temperature gradients in a far-from-equilibrium environment. They could replicate and confine early metabolic and replicative life forms in an abundant aqueous environment.

Surface Characterization of QD-Dopamine Conjugates and Specific Uptake in Mammalian Cells

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In this work, we have used the compound o-phthalaldehyde (OPA) to evaluate the efficiency of dopamine coupling to quantum dots (QDs) via a carbodiimide reaction. The OPA assay is useful for determining the number of dopamine molecules that have bound to the surface of the particles and for measuring the kinetics of the reaction. We find that increasing levels of QD modification leads to changes in the fluorescence spectrum of the particles. Furthermore, when targeted to living cells, these QD-dopamine conjugates show redox specific labeling. In reducing conditions, the QDs are fluorescent mainly in the cell periphery and lysosomal compartments. When the cells are treated to become more oxidizing, the fluorescence spreads throughout the entire cell.

High Aspect Ratio Cantilever Tips for Electrostatic Force Microscopy

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Electrostatic force microscopy is a type of non-contact atomic force microscopy where electrostatic forces are measured. When a conducting tip and sample are used, a capacitor is formed and the capacitance can be measured. A typical commercial tip is pyramidal-like in structure, which leads to contributions from the sides of the tip once the distance is larger than the radius of curvature of the tip. This causes a large background force which can make it difficult to observe small features in spectroscopic curves. We describe a method to make a metallic high aspect ratio tip where these contributions from the sidewalls become minimal.

Birefringent Thin Films of Nanocrystalline Cellulose

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Electrostatic layer-by-layer self-assembled films of nanocrystalline cellulose and poly(allyl)amine hydrochloride (PAH) were prepared by spin-coating and conventional solution dipping. The nanocrystalline cellulose was obtained by acid hydrolysis of cellulose fibres resulting in a stable colloidal suspension of rod-shaped crystals (100-200 nm long by 5-10 nm wide). Surface characterization was performed by AFM, SEM, XPS and reflectance spectrometry. Complete surface coverage was achieved leading to smooth films that displayed high film stability under various conditions due to ionic crosslinking. Samples assembled by spin-coating were substantially thicker, leading to iridescent films after only a few deposition steps. As a novel route to prepare ordered films, nanocrystalline cellulose and PAH were adsorbed from solution in a magnetic field. Optical properties for the various film types were compared and birefringence was measured to determine the orientation of nanocrystalline cellulose in multilayered thin films. Surface order was quantified from AFM data with magnetically aligned films displaying the most order followed by radial order in spin-coated films. These aqueous methods for film preparation present an alternative to iridescent coatings made by chemical vapor deposition, and the anisotropic nature of the system imparts a way to tailor optical properties in thin organic films.

Infrared Nanofocus Maps Sub-10 nm Particles

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Infrared spectroscopy is a powerful tool for material identification based on molecular vibrational fingerprints. However, far-field analysis of individual nanoparticles has not been possible so far. The main obstacle is the extremely weak scattering cross section of nanosize objects at infrared wavelengths, which makes detection impossible in the presence of unavoidable background scattering.

We demonstrate nanoscale resolved infrared imaging of sub-10 nm gold particles by scattering-type scanning near-field microscopy (s-SNOM). Our s-SNOM is basically an AFM, with the metallized probing tip being additionally illuminated by IR light ($\lambda=10 \mu\text{m}$). Due to an optical antenna-effect, light is concentrated at the apex of the tip forming a wavelength independent nanoscale focus [1]. Imaging is performed by scanning the tip over an object placed in the nanogap between the tip and supporting substrate - usually a glass slide. However, in order to make a detection of sub-10 nm particles possible, we employed a simple, but very efficient trick. Instead of using glass as a sample carrier, we replaced it by highly reflecting or polariton resonant materials. That way a strong tip-substrate near-field interaction intensifies the infrared nanofocus illuminating the particles, and the near-field optical particle contrast is enhanced [2].

Our results already promise a wide application potential in high resolution imaging of nanoscale objects (e.g. gold biolabeling). Combined with spectroscopic mapping, our method opens the door to further exciting application: noninvasive, label-free chemical identification of individual nanocrystals or biomolecules via their infrared fingerprint spectra.

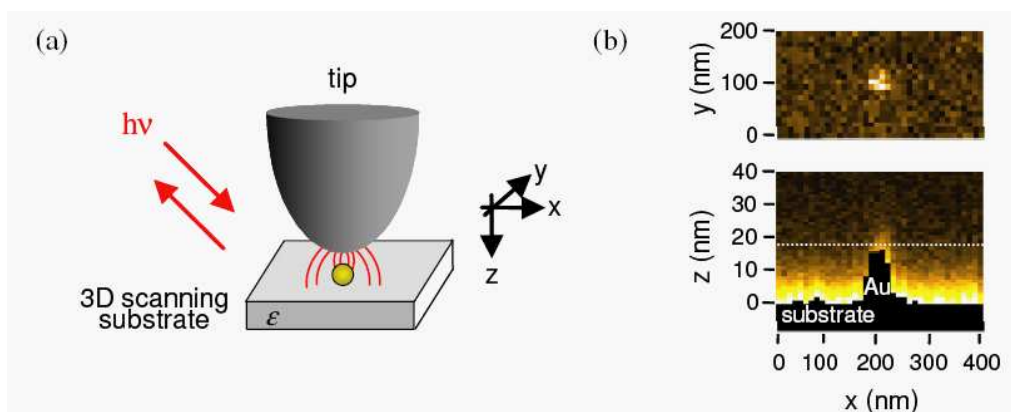


Fig.1 (a) Schematics of the experimental setup. The substrate covered with Au particles is scanned in x , y and z direction and the tip-scattered IR light is recorded at every voxel. (b) Optical amplitude images of 16 nm Au particle on Au substrate. Bottom: vertical (x - z) cross section and Top: horizontal (x - y) cross section (constant height image) above the particle extracted from the 3D data set at a vertical position marked by the dashed white line.

[1] F. Keilmann et al., Phil. Trans. Roy. Soc. A 362, 787 (2004).

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Simulating the Photodissociation of Pyrrole

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Pyrrole provides useful help for the investigation of photochemical processes in the first excited state. The elimination of the nitrogen-bound hydrogen atom takes place, which is observed through quantum chemical simulations with the Restricted Open Shell Kohn Sham formalism [1] using the Car-Parrinello Molecular Dynamics code. Only high kinetic energy hydrogen atoms are obtained [2] that can nicely be illustrated by calculation of velocities at each step of the calculation. Furthermore, effects of surrounding water molecules are expected to play an important role in this photodissociation.

[1] I. Frank et al., *J.Chem.Phys.* 108, 4060 (1998). [2] J. Wei et al., *Faraday Disc.*, 127, 267 (2004).

Anisotropic Deformation Response of Single Protein Molecules

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Single molecule methods have given experimental access to the mechanical properties of single protein molecules. So far, access has been limited to mostly one spatial direction of force application. Here, we report single molecule experiments that explore the mechanical properties of a folded protein structure in precisely controlled directions by applying force to selected amino acid pairs. We investigated the deformation response of green fluorescent protein (GFP) in five selected directions. We found fracture forces widely varying from 100 pN up to 600 pN. We show that straining the GFP structure in one of the five directions induces partial fracture of the protein into a novel, half-folded intermediate structure. From potential widths we estimated directional spring constants of the GFP structure and found values ranging from 1 N/m up to 17 N/m. Our results show that classical continuum mechanics and simple mechanistic models fail to describe the complex mechanics of the GFP protein structure and offer new insights into the mechanical design of protein materials.

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Why Molecules Move Along a Temperature Gradient

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Molecules drift along temperature gradients, an effect termed thermophoresis, Soret-effect or thermodiffusion. In liquids, its theoretical foundation is subject to a long standing debate. Based on a novel all-optical microfluidic fluorescence method, we present experimental results for DNA and polystyrene beads over a large size, salt and temperature range. The data supports a unifying theory based on solvation entropy. Stated in simple terms, the Soret coefficient is given by the negative solvation entropy, divided by kT . Thermodiffusion of polystyrene beads and DNA is predicted without adjustable parameters. The theory is based on assuming local thermodynamic equilibrium of the solvent molecules around the molecule. Such assumption is fulfilled for moderate temperature gradients below a fluctuation criterion. Above, thermodiffusion becomes nonlinear as was shown recently by experiments. For both DNA and polystyrene beads, thermophoretic motion changes sign at lower temperatures. The thermophilicity towards lower temperature is attributed to an increasing positive entropy of hydration, whereas thermophobicity is explained by a temperature insensitive negative entropy background of ionic shielding. The understanding of thermodiffusion sets the stage for detailed probing of solvation properties of colloids and biomolecules. For example, we successfully determine the effective charge of DNA and beads over a size range not accessible with electrophoresis.

New AFM Force Spectroscopy Approach to Analyze the Static and Dynamic Properties of Adsorbed Polymers

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Both, conformation and dynamics of adsorbed polymers govern their behavior and thus their functionality. On the one hand, the end-to-end distance and the contour length of single carboxy-methylamylose (CMA) polymers were determined with a mechanical approach. The polymer chains, covalently pinned with one segment to the surface, were picked up randomly with the AFM tip. The distance between pickup-point and pinpoint corresponds to the end-to-end distance of this arbitrary polymer part. The location of the pinpoint was derived by measuring the normal force while laterally scanning the surface at constant height (see Fig. 1). Due to the self-similarity of the polymer, the scaling exponent of CMA was found to be 0.744 ± 0.04 , which indicates that the polymer is mobile and rearranges on the surface while absorbing on it. Rare crossovers or polymer loops may occur.

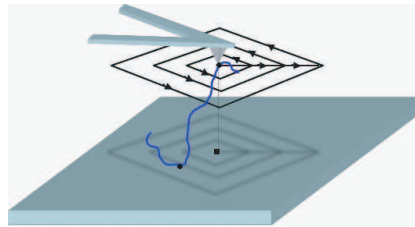


Fig.1 AFM procedure to determine the end-to-end distance of carboxymethylamylose. After picking up the polymer, the tip is scanned parallel to the surface in a certain height in squares with increasing diameter. Thereby, the pinpoint and thus the end-to-end distance to the pickup-point can be determined from the measured normal force spectrum.

On the other hand, the mobility and frictional behavior of polymers like ssDNA and polyallylamine (PAAm) were investigated by new AFM measurement protocols (see Fig. 2). The polymer chains were covalently attached to the AFM tip and were allowed to adsorb on a mica or gold surface. During the desorption process, the tip was additionally moved parallel to the surface, so that a horizontal force component induced a sliding of the adsorbed polymer part depending on the friction coefficient. Thus, the obtained force-extension spectra reveal detailed information on the mobility and friction of a polymer chain on a surface under experimentally accessible conditions. Furthermore, the AFM allows a spatially resolved determination of the friction coefficient on the basis of the interaction of individual polymer molecules.

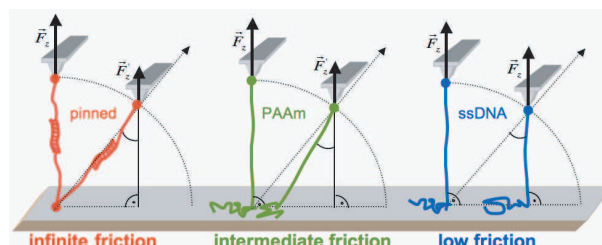


Fig.2 Geometric schemes of the experimental procedures to investigate three different friction force regimes. The desorption process occurs with an angle to the vertical direction. The resulting horizontal force component induces a sliding of the adsorbed polymer part depending on the friction coefficient.

Laser Cooling of Micro and Nanomechanical Levers

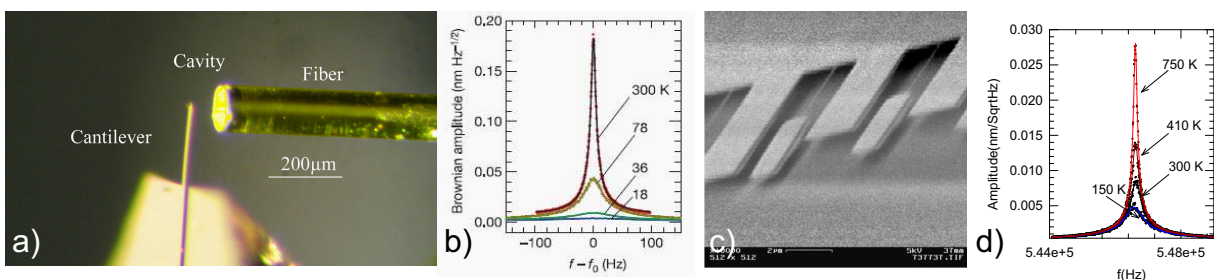
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The prospect of realizing entangled quantum states between macroscopic objects and photons has recently stimulated interest in new laser-cooling schemes of macroscopic mechanical resonators, ranging from millimetric mirrors to microscopic levers. For instance, laser-cooling of the vibrational modes of a mirror can be achieved by subjecting it to a photon-induced force, actively controlled through a feedback loop in order to oppose the mirror's Brownian motion [1]. In contrast, atoms can be laser-cooled passively without having to make use of such an active feedback, because their Brownian motion is intrinsically damped through their interaction with photons in a standing wave thanks to the Doppler effect (Doppler cooling of atoms) [2]. Here we present the new principles and recent experimental studies of different passive laser-cooling schemes of micro and nanomechanical resonators. All our schemes rely on a time-delayed optomechanical coupling between the motion of the resonator and the photon field of an optical cavity.

In a first scheme, we address the cooling of a micromechanical resonator being used as one of the two mirrors of a Fabry-Perot optical cavity (Fig. 1a) [3]. Using photothermal effects, we are able to quench passively the Brownian vibrational fluctuations of a gold-coated silicon AFM microlever (7 kHz) from room temperature down to an effective temperature of 18 K (Fig. 1b) [3]. This photothermal cooling scheme also enables us to cool from room temperature down to 150 K the vibration mode of high frequency mechanical resonators (600 kHz) of a much smaller size (Fig. 1c and 1d). In order to reach the quantum ground state of such vibrational mode, it is desirable to avoid residual optical absorption. For this purpose, this cooling scheme could favor the use of radiation pressure over light induced photothermal effects [3,4,5].

We then present the principles of a new cooling scheme for nanomechanical resonators. By placing a nanomechanical lever in the field of a high finesse optical cavity, the mechanical motion of the lever is coupled to the dynamics of the photon-field in the cavity, through scattering of the photons by the nanolever. This optomechanical coupling, delayed by the response time of the cavity, allows cooling of the lever vibration mode. This new scheme, as opposed to the previous one, can address objects with size smaller than the diffraction limit, as nanomechanical resonators with vibration frequencies in the GHz range. It also allows the study of their mechanical properties.



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[4] S. Gigan et al, quant-ph/0607068 (2006).

[5] O. Arcizet et al., quant-ph/0607068 (2006).

Single Molecule Studies of Eukaryotic Transcription using Optical Tweezers

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Our goal is to investigate the behavior of single RNA Polymerase II molecules during the DNA transcription process. In detail we are interested in the molecular mechanisms of eukaryotic transcription especially, the complex behaviour of the elongation process, changes in transcription velocities and effects of transcription factors. Here, we show the experimental setup of dual beam tweezers, and first data. Experimental data together with simulated noisy data are used to illustrate challenges in the analysis.

pH-Sensitive Shielding Strategy for Polycation Based Non-Viral Gene Delivery Systems

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For shielding polyethylenimine/DNA polyplexes against unspecific interactions, surface amines can be reacted with activated ester of PEG. N-hydroxylsuccinimide-pyridyl-hydrazone-PEG (mPEG-HZN-NHS) was applied for pH-sensitive reversible shielding. Polyplexes were generated with a ligand for cell targeting, and reacted with mPEG-HZN-NHS for 1 h. Particles were then analyzed with regard to their biophysical properties and gene transfer efficiency in vitro and in vivo. Polyplexes of around 200 - 400 nm with a neutral surface charge (zeta potential of less than +5 mV) were generated. After 1 h incubation at 37°C at pH 7.4, particles remained shielded, whereas at pH 5 particles were deshielded. Luciferase gene expression showed that the targeted polyplexes with reversible shield exhibited up to 30-fold greater gene expression compared to stable-shielded control polyplexes.

Targeting of tumor cells after tail vein injection into tumor bearing mice was achieved by introducing a PEG coupled targeting ligand.

Switchable Aptamers and Transcriptional RNA Circuits

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Aptamers are oligonucleotides that bind to specific target molecules, such as proteins. Making them switchable means changing their conformation from a target binding to a non-binding conformational state and vice versa. This conformational change can be achieved by alternatively adding suitable oligonucleotide counterstrands. Under certain conditions, an oscillating RNA concentrations in in vitro transcriptional circuit can be achieved. If one of the counterstrands involved in the aptamer switching is such an RNA species, its concentration oscillation could autonomously drive the switching of the aptamer.

Transport Through Quantum Dots in a Magnetic Field

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Using the method of infinitesimal unitary transformation (flow equations), we derive a perturbative scaling picture of the Non-Equilibrium Transport through a Kondo Dot in a Magnetic Field. The scaling picture generically contains both the equilibrium and the non-equilibrium physics of the model, thereby allowing us to investigate the effect of the magnetic field on the system using a consistent description for zero and nonzero dc-voltage bias.

Structural Studies on RNA Polymerase I

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RNA polymerase I (Pol I) transcribes ribosomal DNA (rDNA) units, which are found in multiple clustered copies in every eukaryotic cell. After processing of the ribosomal RNA (rRNA) precursor, the mature RNA is subsequently incorporated into nascent ribosomes. Pol I consists of 14 polypeptides, 5 of them are shared with RNA polymerase II and III, 7 peptides show homology based on the primary sequence and 2 of them (A49 and A34.5) are unique subunits of Pol I. The A49/A34.5 subcomplex together with A43 and A14 are responsible for the enzymatic specificity of rRNA transcription by Pol I. In order to understand the specific transcription mechanism of Pol I, more structural information is needed. The aim of our work is to establish the 3-D structure of Pol I. For this purpose two complementary strategies are pursued. First the endogenous protein complex (approx. 600 kDa) from yeast was purified and we try to elucidate the total structure at medium resolution. The second approach is the recombinant expression, purification and crystallization of the A49/A34.5 and A43/A14 subcomplexes of Pol I. The results obtained will give more detailed structural information, which will lead to a better understanding of the specific transcription process of Pol I.

Bimodal AFM-Imaging of PS-PB Diblock-Copolymer Surfaces

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To map material contrast to topographical features is one of the most common applications in Scanning Probe Microscopies. However, the information of these two cannot be determined independently. Topographical crosstalk into material channels such as the phase contrast in resonant tapping mode imaging has to be avoided in order to quantify the real physical contrast. In this contribution we present a new measurement technique, that was devised to alleviate this problem, the so called bimodal approach. In this mode of operation, both the first and the second eigenmodes of the cantilever resonances are excited mechanically. While the first eigenmode is only used in order to remain in close contact with the surface, the second eigenmode amplitude and phase signal is only minimally influenced by the topography of the sample. First results on a self-assembled surface of polystyrene-*block*-polybutadiene diblock copolymer using this technique will be shown.

Nanoscale Optical Imaging of Carbon Nanotubes

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Optical techniques with nanoscale spatial resolution are essential for the detection and analysis of individual nanoobjects such as single molecules, semiconductor quantum structures and biological proteins. We apply a near-field optical technique that allows to perform photoluminescence (PL) and Raman spectroscopy with 10 nm spatial resolution on the same nanostructure and use it to study single-walled carbon nanotubes (SWCNTs) [1]. The technique is based on the field enhancement effect at sharp metal structures. A laser-irradiated metal tip localizes and enhances the optical fields at its apex thereby creating a nanoscale light source (Fig. 1). The metal tip is brought in close proximity to the sample surface and Raman scattered light or PL light is collected and assigned to the momentary tip position. By raster scanning the tip, a high-resolution, chemically specific map of the sample surface can be recorded.

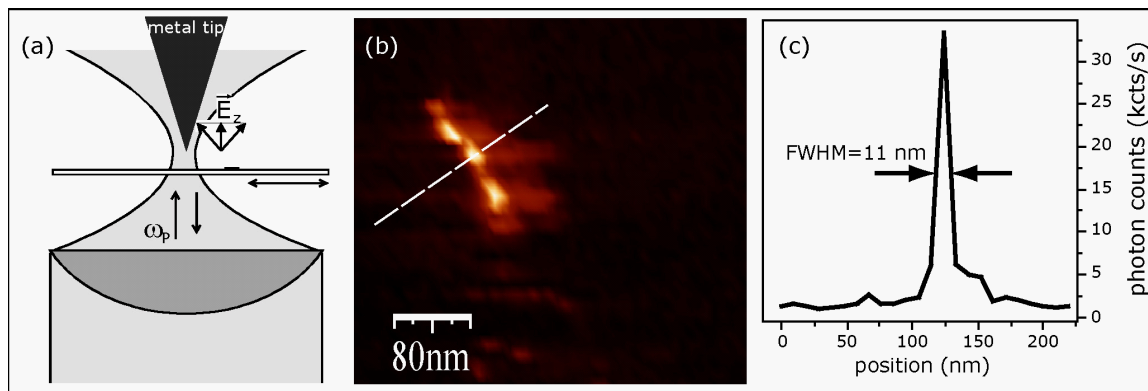


Fig.1 (a) Schematic of the experimental setup. (b) Near-field PL image of a SWCNT on glass. The PL image is formed by detecting the PL intensity around 950 nm after laser excitation at 632.8 nm. (c) Cross section taken along the dashed line in (b).

We present near-field PL and Raman imaging of SWCNTs with a spatial resolution of about 10 nm. Highly localized excitation is used to visualize the spatial extent and energy of the contributing excited states [2]. We show that the emission can occur in confined segments along individual tubes for our samples. Signal enhancements achieved for Raman and PL on the same tube are quantified and different contributions are discussed.

[1] A. Hartschuh et al., Phys. Rev. Lett. 90, 095503 (2003). [2] A. Hartschuh et al., Nano Lett. 5, 2310 (2005).

Binding Site Water Carries Information About the Binding Site

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Many proteins function by the formation of a transient complex with a protein partner. These proteins have to choose with high reliability between a plethora of possible interaction partners. Thus, the information about which proteins to choose as a partner needs to be encoded in the proteins architecture and to be transmitted through space. We are interested in the question if this information is transferred to the surrounding solvent. Here, we have quantified the information content of the solvent with the Shannon entropy definition. We can show that the arrangement of water molecules has a higher information content near binding sites than elsewhere. This suggests that water carries information about the binding

sites. The information content of the binding-site water can help in mediating specific interactions in early stages of the complexation process. The Shannon definition of entropy is closely related to the Gibbs entropy definition derived from statistical thermodynamics, implying that the higher information content may be related to lower thermodynamic entropy of binding site water. Therefore, one can assume that the specific interaction of proteins is facilitated by an entropy gain of the solvent. This is conceptually related to the hydrophobic effect in protein folding, even though binding sites of transient protein complexes are not more hydrophobic than the average protein surface.

Supramolecular Assemblies as Template Structures for the Manipulation of Nano-Scale Objects

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A cheap and easy to use ambient condition method based on a two-dimensional molecular organic template acting as a host architecture for the defined fixation of guest molecules is presented. We report on a new method for the investigation and manipulation of single molecules by STM without the explicit need for experimentally challenging UHV techniques. The controlled manipulation of single molecules becomes possible, since an organic template preadsorbed on the substrate introduces a hexagonal grid of stable adsorption sites with a next nearest neighbor distance of approximately 1.6 nm.

Therefore the adsorption of Trimesic Acid (TMA) to various single crystal surfaces has been studied under Ultra High Vacuum and ambient conditions. The self-assembled structure is characterized by periodic non-dense-packing of the molecules. Depending on the preparation method, in UHV as well as in ambient conditions, two different network structures could be realized. In both phases, induced by directed hydrogen bonding, the organic molecules built a two-dimensional grid architecture with molecular caves – both able to store guest molecules at specified adsorption sites. On the liquid solid interface, it was possible to adjust one of the two polymorphs by choosing the suitable solvent whereas in UHV preparation parameters are more crucial.

After preparation of the TMA host structure on a crystal surface TMA molecules themselves were inserted as guest molecules into the host structure. The guest molecules could be identified in two different vertical and 6 different horizontal adsorption sites. In the horizontal case STM induced switching of a single guest molecule to six stable positions was observed. The states of this molecular switch have a distance of 0.15 nm. The calculated energy barriers indicating that the switch is stable at room temperature are consistent with the experiment.

Furthermore Buckyballs could be inserted in the hollow sites. Buckyballs have been shown to be suitable for translational manipulation with the STM, but so far mostly under UHV conditions. With a diameter of about 0.7 nm C₆₀ fits easily in the pores of the TMA structure which have a diameter of about 1 nm. By means of STM the Bucky-Balls could be imaged within the template structure and be directly kicked from one cell to another.

As a third example coronene molecules were co-crystallized after the host structure was formed. With a diameter of about 1 nm coronene molecules also fit quite well into the open pores of the host. Closed layers of TMA with every cavity filled by a coronene molecule could be imaged. Even the submolecular structure of coronene can be seen in the STM images. The STM topographs suggest still standing and rotating coronene molecules – mediated by a template induced variation of the adsorption site of the coronene molecules on the graphite surface. By applying voltage pulses the guests molecules could controllable be kicked out of the cavities.

Observing and Controlling Single-Molecule Enzyme Kinetics

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Understanding the mechanisms underlying enzymatic catalysis on the single-molecule level is a key issue on the way to creating artificial Nanobiomachines.

Lipases and esterases are exoenzymes that belong to the group of hydrolases, which are generally extremely stable and therefore widely used in industrial applications. Here, the lipase B from *Candida antarctica* (CalB) is utilized for catalyzing a saponification reaction that converts the non-fluorescent substrate carboxyfluorescein ester (CFDA) into the highly fluorescent product carboxyfluorescein which can be observed, reporting the completion of one enzymatic turnover cycle. Single CalB molecules were covalently bound to a glass cover slip and imaged at approx. 200 Hz by wide-field TIRF microscopy using an EMCCD camera. Hence the kinetics of catalysis can be derived from statistical analysis of the fluorescence timetraces of each individual molecule. Simultaneous manipulation of the system is achieved by a functionalized microbead bound to an AFM cantilever. Pulling enzymes using a specific antibody with known binding rate leads to conformational changes of the enzyme that influence the reaction coordinate of the catalytic reaction.

Surface Reactions on Functional Self-Assembled Monolayers

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Self-assembled monolayers have many potential applications in modern technology [1,2]. Uses of self-assembled monolayers range from the control of properties of surfaces (adhesion, wettability, etc.) to the fabrication of new materials for sensors and nonlinear optical devices. Development in these fields depends upon the availability of self-assembled monolayers bearing reactive functional groups at their surfaces. These reactive functional groups are necessary to elaborate monolayers into multilayer films (for new materials) or anchor molecular fragments to the monolayer surface for device applications.

In-situ modification of a suitable precursor monolayer where the anchor groups are already bound to the surface can be attempted [3]. This approach often leads to problems because the quantitative conversion of a surface functionality is not trivial due to the rigid, densely packed environment of a surface-bound monolayer. Therefore, different criteria exist for chemical reactions which can be used on a surface: (a) the quantitative conversion under mild conditions, (b) the compatibility with different functional groups and (c) the absence of byproducts, which might accumulate at the interface and retard or stop the surface reaction. These criteria are closely related to the click chemistry concept which was introduced by Sharpless [4]. The goal of this approach is to develop an expanding set of powerful, selective, and modular “blocks” that work reliably in both small- and large-scale applications.

The most perfect click reaction known to date is the Huisgen 1,3-dipolar cycloaddition of azides and acetylenes [3]. This cycloaddition is irreversible and proceeds in quantitative yield without any side products in many solvents. Azide and acetylene groups are tolerant to many other functionalities.

In this contribution we will present the modification of silica substrates by using the copper catalyzed 1,3-dipolar cycloaddition of azide-bound monolayers with different acetylenes. As acetylenes we choose a coumarin dye with a terminal triple bond for the click reaction. Successful surface modification was characterized by IR spectroscopy, XPS and contact angle measurements.

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[3] T. Lummersdorfer et al., *J. Phys. Chem B*, 108, 3963 (2004). [4] H. C. Kolb et al., *Angew. Chem. Int. Ed.* 2004, 40 (2001).

Photophysical Properties of Deep Blue Emitting Iridium Complexes

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Close to a quarter of electrical power generated is used to feed artificial light sources. Improving the efficiency of lighting, which still evolves around the incandescent bulb, is therefore imperative in optimizing the use of available energy resources. Electrophosphorescence based organic light-emitting diodes (OLEDs) have shown an internal efficiency of nearly 100% [1].

This is achieved through the harvesting of both singlet and triplet excitons generated by the charge injection process at the electrodes. For lighting applications it is mandatory to produce highly efficient OLEDs with white light emission. For this purpose, device architectures based on red, green and blue phosphors have been proposed.

Considerable interest is focused on Iridium(III) metalorganic complexes as phosphorescent emitters. The main advantage of these materials lies in the wide range of colour tunability, depending on the chemical structure, and the high phosphorescence quantum efficiencies achievable due to the large spin-orbit coupling. Highly efficient green and red OLEDs based on these complexes have been successfully demonstrated. However, the development of blue emitting phosphors with reasonable photophysical stability and operational lifetime is a real challenge and still needs to be addressed.

Here we present results on a new family of phosphorescent Iridium carbene complexes [2] emitting in the blue, which are relevant for the design of blue phosphorescent emitters and the development of white light OLEDs. By a combination of steady-state and time-resolved spectroscopy techniques we study the photophysics of the complexes in thin films and in solution. The results demonstrate the influence of the ligand chemical structure in tuning the emission colour. Moreover, the nature of the non-radiative pathways involved in the emission process is investigated on the basis of phosphorescence lifetime and quantum yield measurements. These non-radiative pathways are important in limiting the overall device efficiency.

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[2] BASF AG, PCT Patent Application WO 2005/019373 (2003);

Universal Display Corp., PCT Patent Application WO 2005/113704 (2004).

Attaching Fréchet Dendrons - a Route to Constructing Functional Supramolecular Assemblies on a Graphite Surface

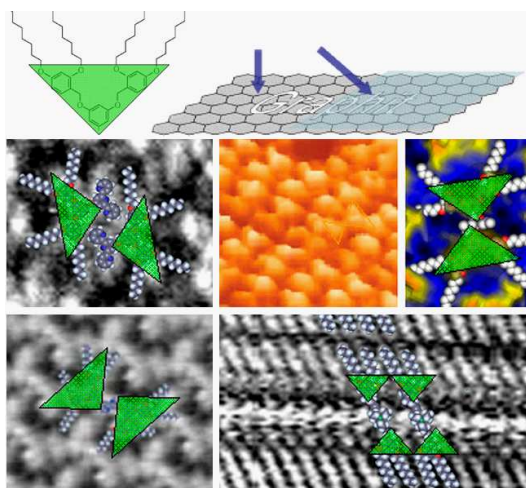
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Self-organised molecular layers provide a promising route to constructing new (switchable) nanodevices and optimizing nanosensors. The design of such molecules involves control of structure and stabilization on the surface as well as control of functionality. The search for a potentially universal “adapter” which can control the self-organization and stabilization on a particular type of surface is a unique challenge. High-quality nanoscale imaging using scanning tunneling microscopy (STM) provides the means to further such studies. We present here a review on our recent STM work on self-organized monolayers of a general “self-organizer” for graphite surfaces, i.e. a Fréchet-type dendron. How powerfully this self-organizing motif can affect various central components (catalytic, switchable, redox-active) on a graphite surface is addressed by analyzing self-organized monolayers of nine different molecules, each containing at least one first or second-generation Fréchet-type dendron.



Tube Radius in Entangled Networks of Semiflexible Polymers

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The mechanical properties of the cytoskeleton play an important role in many cellular functions like locomotion or adhesion. One of the cytoskeleton’s dominant constituents is a network structure composed of the semiflexible polymer F-Actin. To connect the single polymer properties to the macroscopic behavior of the network, a single polymer is considered to be constrained to a tube established by neighboring filaments. This tube concept can then be used to obtain a variety of mechanical properties of the complete network. While scaling laws for the tube diameter are well established, the absolute value is still under debate and different theoretical concepts and experimental measurements exist.

We present a new approach to the problem and have conducted extensive computer simulations to check the validity of our assumptions. A model of independent rods is used to describe the confinement of a single semi-flexible polymer in the network environment. A self-consistency approach allows then to derive an absolute tube radius for the network as a function of several parameters.

Spin Related Transport Phenomena in One-Dimensional Electron Systems

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We investigate spin related transport phenomena in quasi one-dimensional wires which are lithographically defined in semiconductor quantum wells. We explore electron spin effects in such mesoscopic structures either by exploiting the Zeeman coupling of the electron spin to an external magnetic field or by utilizing spin-split edge states of a Landau quantized two-dimensional electron gas as source and drain contacts. Furthermore, by applying a voltage to a close-by gate we can adjust the energy of the one-dimensional subbands relative to the Fermi energy of the two-dimensional reservoirs. The main focus of our experiments is to optically manipulate the spin-polarized electron transport through the one-dimensional quantum wires. To this end, the quantum wires are fabricated by a combination of optical and electron beam lithography; enabling large one-dimensional subband energies of about 10 meV, while the lateral widths of the quantum wires are typically in the order of 100 nm. First transport experiments at 4.2 K nicely demonstrate the spin-splitting of the one-dimensional subbands at a magnetic field of about 5 Tesla.

Spin Relaxation in n-InGaAs Wires: Transition from 2D to 1D

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For an efficient information processing scheme based upon the electron spin, it is important to explore carrier spin relaxation mechanisms in nanostructures as a function of dimensionality. In two and three dimensions, elementary rotations do not commute, with significant impact on the spin dynamics if the spin precession is induced by spin-orbit coupling. Spin-orbit coupling creates a randomizing momentum-dependent effective magnetic field; the corresponding relaxation process is known as the D'yakonov-Perel' mechanism. In an ideal one-dimensional system, however, all spin rotations are limited to a single axis, and the spin rotation operators commute. In the regime approaching the one-dimensional limit, a progressive slowing and finally a suppression of the D'yakonov-Perel' spin relaxation have been predicted.

We report on spin dynamics of electrons in narrow two-dimensional n-InGaAs channels as a function of the wire width [1]. We find that electron-spin relaxation times increase with decreasing channel width, in accordance with recent theoretical predictions. Surprisingly, the suppression of the spin relaxation rate can be detected for widths that are an order of magnitude larger than the electron mean free path. We find the spin diffusion length and the wire width to be the relevant length scales for explaining the observed effects.

We acknowledge financial support by AFOSR, ONR and CeNS.

[1] A.W. Holleitner et al., Phys. Rev. Lett. 97, 036805 (2006).

Investigation on the Geometry and Mobility Dependence of the Quantum Hall Effect

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On the contrary to the usual localization and Landauer type edge state theories, the self-consistent screening calculations predict that the quantum Hall plateaus strongly depend both on the sample width and mobility. In this work, we investigate experimentally the formation of these plateaus considering surface gate defined Hall bars in the quantum Hall regime at low (< 4 K) and high (> 15 K) temperatures for various samples sizes ($1 \text{ micron} < w < 10 \text{ micron}$) and mobilities ($\mu = 1.8, 2.5$ and $10 \text{ million cm}^2/\text{Vs}$). The outcome of temperature dependency of the activation is also decisive to conclude the relevancy of the mentioned theories at narrow samples, which we measured at predicted T values.

Single Molecule Studies of Proteasome Complexes Using Combined Fluorescence and Atomic Force Microscopy

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The 20S proteasome is a large 700kD protein that is involved in the degradation of misfolded proteins as well as in the degradation and processing of short-lived regulatory proteins [1]. It has a height of 15 nm, a diameter of 11 nm and a barrel-shaped body, which is traversed by a channel that widens into 3 inner cavities. The central cavity harbours the active sites and the two outer chambers are putatively involved in substrate translocation. We aim to gather information on the occupation of these chambers by substrate proteins in order to better understand the translocation process.

We study complexes formed by folded substrate proteins trapped inside the chambers of irreversibly inhibited proteasomes. Proteasomes and substrate proteins can be fluorescently labelled by different fluorophores. We use combined confocal microscopy and AFM to get a “snapshot” of the translocation process on a single molecule level. The sample is raster scanned by a cantilever from above and is simultaneously illuminated by an excitation laser from the bottom through an inverted optical microscope. Topographic images allow us to resolve individual proteasomes on a cleaned glass cover. The fluorescence from up-to two different fluorophore species is separated and detected in two different detector channels. Fluorescence provides correlation information on the occupation of the proteasomes. Additionally, the distance between two labelled proteins within separate chambers or between the proteasome central chamber and the substrate protein can be estimated by the occurrence of FRET.

Further experiments will improve optical resolution by using SNOM with aperture cantilevers and tip-on-aperture cantilevers [2]. Thus we can image highly dense samples and may be able to gain additional information on the proteasome-complex substructure by direct imaging.

[1] W. Baumeister et al., Cell Vol. 92, 367 (1998). [2] H. Frey et al., Phys. Rev. Lett. 93, 200801 (2004).

Molecular Simulations of DNA Oligonucleotides

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Studies of synthetic oligodesoxynucleotides (ODN) expressing unmethylated CpG motifs show that these molecules have immuno-regulatory effects, being recognized as a pathogen-associated molecular pattern triggering a rapid immune response. Structural analysis of interaction between CpG motifs and Toll-like receptors leads to a better understanding of how the mammalian immune system has evolved to selectively recognize pathogens, giving insights for the rational use of DNA technology in DNA vaccines, gene therapy and in anti-cancer drug design. Using model building and molecular mechanics studies we were able to construct the tertiary structure of a synthetic CpG oligonucleotide. A 3D model of the double-helix conformation was constructed based on our secondary structure analysis in combination with previous experimental results. After 6 ns dynamic simulation structural information about the behavior of this molecule was obtained. The stability of the molecule was shown and a detailed structural analysis was performed.

Diffusion of Organic Molecules in Mesoporous Silica Materials

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The understanding of molecular dynamics in confined nanometer-sized geometries is one of the most important aspects in current research aimed to design novel smart materials based on host/guest systems such as drug-delivery materials. We investigate the translational, orientational and spectral diffusion of organic molecules in nanoporous silica materials using confocal and widefield fluorescence microscopy. The mesoporous channel system could be thought of a system of pipes in which molecules can diffuse. Analyzing the trajectories of the guest molecules by single particle tracking allows us to characterize the influence of the host system on molecular diffusion.

Two different studies are presented. First, we investigate simultaneously orientational-translational and orientational-spectral dynamics of single terrylendiimide (TDI) dye molecules as guests in the pores of an M41S host silica film synthesized with the ionic ammonium surfactant CTAB as template. Using confocal microscopy the in-plane angle of an individual molecule can be obtained with a precision of a few degrees, and can be correlated with the corresponding spectrum. Moreover, measuring simultaneously the position and the orientation of TDI shows molecules moving back and forth within the pore architecture. The movement is extremely linear, with the overall distance covered $\approx 1 \mu\text{m}$.

In a further step, short interfering RNA (siRNA) strands labelled with Atto 647N are incorporated into the pores of a thin M41S-film synthesized with the non-ionic Block-copolymer Brij-56 as template. RNA interference methods using siRNA represent a very promising strategy in the therapy of various different cancers. The diffusion of the biomolecules within the pore-system can be detected using widefield-microscopy. The understanding of the dynamics of the siRNA molecules in such mesoporous films is an essential prerequisite for their incorporation into novel silica drug-delivery systems.

Coulomb Repulsion Effects in AC-Driven Molecular Wires

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We investigate within a master equation approach the effects of strong Coulomb interaction in molecular wires. As compared to non-interacting electrons we find modified length-dependencies and scaling laws. We generalize our approach to the case of a molecular wire under the influence of an AC field.

Hydrogen-bonded Molecular Networks at the Liquid-Solid Interface

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Molecular monolayers were grown by spontaneous self-assembly from solution at the liquid-solid interface and subsequently investigated by Scanning Tunneling Microscopy (STM). The goal is to get a more profound understanding of the driving forces, dependence on external parameters and conditions that result in self-assembly of interfacial monolayers. We present co-adsorption of two different molecules, BTB (1,3,5-benzenetribenzoic acid) and TMA (trimesic acid) in open (loosely packed) networks, studied in two different solvents (heptanoic and nonanoic acid). Altering the absolute and relative concentrations of the two compounds in binary solutions resulted in six mixed phases with different structures and stoichiometries. All structures are stabilized by twofold intermolecular hydrogen bonds between the carboxylic acid head groups. Moreover, in-situ dilution of the supernatant solution induced phase transitions of the monolayer structures, accompanied by an alteration of the size and shape of cavity voids in the 2-dimensional molecular assembly. Furthermore, the dynamical behavior of various dicarboxylic acid molecules was investigated at the liquid solid interface by comparison of structurally related compounds. The three molecules NDA (2,6-Naphthalenedicarboxylic acid), BPDA (4,4'-Biphenyldicarboxylic acid), and SDA (4,4'-Stilbenedicarboxylic acid) have two interconnected benzoic acid groups in common, however the spacing varies. For NDA the two benzoic acid groups are linked through a π -bond, whereas for BPDA and SDA they are separated by spacers of variable length. It could be shown, that the dynamic behavior, e.g. island growth and ripening, of the monolayer structures is not dominated by the size, shape and binding properties of the molecules but by the environment, namely the surrounding solution.

Chemical Aspects of Place Exchange Reactions

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Gold nanoparticles (AuNPs) are being studied/developed for applications in biosensors, drug-delivery and optically active materials. To create versatile particles, researchers mainly rely on place exchange reactions (PERs). These reactions which involve the substitution of a surface bound thiol with a solution thiol have been studied for over 10 years; however their mechanism is still under debate. PERs were studied using HPLC, GC, TEM, NMR and UV-Vis spectroscopy. The results of combining these techniques enable a coherent picture of the kinetics of PERs to be developed and to link the mechanism of 2-D and 3-D PERs.

Nonequilibrium Kondo Physics: Voltage Bias and Time Dependence

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The flow equation approach [1] is a new nonperturbative analytical approach to quantum many-particle systems based on a sequence of infinitesimal unitary transformations that diagonalizes the Hamiltonian. The full Hilbert space is retained in this procedure, which makes this method particularly suited for studying nonequilibrium problems. I will discuss the application of the flow equation approach to the nonequilibrium Kondo model, which is of paradigmatic importance for correlated nanostructures. In particular for the case with large voltage bias (i.e. beyond the linear response regime) physical observables like the dynamical and static spin susceptibility give insights into the nonequilibrium steady state and allow to compare it with finite temperature equilibrium states.

[1] S. Kehrein, *The Flow Equation Approach to Many-Particle Systems* (Springer Verlag 2006).

Synthesis of Magnetic Nanowires in Low-Dimensional Alumina Templates

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Regular arrays of metallic and magnetic nanowires have a vast number of potential applications, for example in radiation and magnetic sensing, high-density magnetic devices, high-density storage media, functional nanomaterials exhibiting quantum size effects, highly sensitive chemical sensors, nano-electronic devices and functional biochemical membranes [1]. Employing anodic alumina membranes (AAM) for nanowire growth is a widely used method to produce highly ordered nanowire arrays at low cost, with a high yield and a high throughput. Alumina templates provide a remarkable hardness, uniform pore size and high pore density [2].

Co and Ni nanowires were synthesized in both commercial and self-prepared AAMs by electrochemical deposition and characterized by scanning electron microscopy (SEM), SQUID magnetization, and electron spin resonance (ESR) experiments. The observed Curie temperatures agree well with those of bulk material and the hysteresis indicates soft ferromagnetic behavior. ESR exhibits a pronounced dependence on the shape and orientation of the sample. Ni self-prepared samples with higher pore density show a thin-plate like anisotropy (i.e. hard axis perpendicular to the plane), indicating magnetic interaction between the wires, while in the 200 nm diameter commercial samples the wire-like (i.e. easy axis along the wire) and the thin-plate like behavior compete.

[1] M. Zacharias et al., P. Physik Journal 4, 29 (2005).

[2] G. E. Thompson et al., Thin Solid Films 297, 192 (1997).

Synthesis and Characterization of Mesoporous Palladium/Tin Oxide for Sensor Applications

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Semiconducting tin oxides are of great interest for sensing applications with respect to hydrogen and carbon monoxide [1]. Their sensitivity can be enhanced by doping, e.g. palladium or by introducing mesoporosity for faster access to active sites. The aim of this work was to combine both ideas and to synthesize and characterize mesoporous palladium containing tin oxides.

Thin films were deposited on cleaned silicon and glass substrates using sol-gel and spin-coating techniques [2]. The films were aged and calcined for the formation of crystalline nanoparticles. Characterization was carried out by X-Ray Diffraction (XRD), Scanning Electron Microscopy (SEM), Energy Dispersive X-ray spectroscopy (EDX), Transmission Electron Microscopy (TEM), and Rutherford Backscattering Spectroscopy (RBS).

Wormlike mesostructures were found with crystalline tin oxide nanoparticles with approximately 5 nm in diameter. The thin films are semiconducting and of high interest for sensor applications which will be investigated in the future.

[1] G. De et al., Sensors and Actuators B 55, 134 (1999), [2] Y. Lu, R. Ganguli et al., Nature 389, 364 (1997).

In-situ GISAXS Study of the Formation/Phase Transition of Silica Mesostructures within Anodic Alumina Membranes

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The synthesis of mesostructured material within the pores of anodic alumina membranes (AAM) has been established recently [1]. The phase of the mesostructure formed during the evaporation induced self assembly (EISA [2]) is strongly depending on the nature and concentration of the structure director and on the synthesis conditions, especially humidity and temperature. With respect to the latter the formation mechanism of the mesostructure within the confined environment of the AAM channels was investigated by in-situ grazing incidence small-angle X ray scattering (GISAXS) experiments.

A columnar and circular hexagonal and a lamellar structure were observed. The phase transformation of a circular hexagonal structure into the columnar hexagonal or a mixture of the columnar hexagonal and the lamellar phase were observed. The existence of a postulated lamellar phase was proven by GISAXS and TEM investigations [3]. The prediction [1] of the kinetically favoured formation of the circular hexagonal structure over the thermodynamically stable columnar hexagonal structure was strongly supported in this work.

[1] B. Platschek et al., *Angew. Chem., Int. Ed.* 45, 1134 (2006). [2] C. J. Brinker et al., *Adv. Mat.* 11, 579 (1999). [3] Y. Wu et al., *Nature Materials* 3, 816 (2004).

Storage of Excitons in Elongated Nanocrystals

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Spherical CdSe nanocrystals capped by a CdS rod-like shell, referred to as nanorods, exhibit interesting spectral dynamics on the single particle level [1,2]. However, for the purpose of applications, the ensemble properties of nanorods are most interesting. We are especially interested in the behaviour of an ensemble of nanorods under the influence of an electric field, as this holds great relevance for future devices. We show here that by applying an electric field to an ensemble of nanorods in a vertical sample geometry fluorescence quenching coincides with a suppression of radiative rate without increasing ionization and non-radiative decay. After turning off the electric field, a significant fraction of quenched – and therefore stored - excitons recombines radiatively, even for field lengths up to 100 μ s. Despite the reduction of the wavefunction overlap arising from the field induced separation of electron and hole the coulombic binding of the exciton prevents enhanced trapping of carriers on the highly reactive surface or in potential fluctuations in the nanocrystal lattice. As exciton storage selects the most polarizable particles, a significant quantum confined Stark shift of ≈ 15 meV along with a correlated broadening of the spectrum is observed in the time-resolved emission of the ensemble at room temperature. This controlled modulation of both the emission wavelength and width together with the dramatic extension of the nanoparticle radiative lifetime opens up promising applications in optical modulators and optical memory cells.

[1] J. Müller et al., *Phys. Rev. Lett.* 93, 167402 (2004). [2] J. Müller et al., *Nano Lett.* 5, 2044 (2005).

Adsorbates on TiO₂(110) - What Can Low Temperature STM Experiments Add?

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Cryogenic temperatures and a UHV environment provide the conditions for stable STM operation, facilitating high resolution imaging, local spectroscopy, and single atom / molecule manipulation. Combination of these experimental capabilities was utilized to study the interaction of adsorbates with oxygen vacancies on TiO₂(110). This type of point defect is common for reducible transition metal oxides and greatly influences their surface chemistry.

Since their unexpected catalytic activity was discovered, Au-nanoparticles on TiO₂(110) have received considerable attention. Deposition of Au at low temperature inhibits thermally activated diffusion and aggregation, thus leads to adsorption of single Au-atoms. Two different distinctive adsorption sites have been identified through STM imaging and manipulation: Au-monomers either adsorb on bridge sites between five-fold coordinated Ti atoms or in oxygen vacancies, i.e. substituting the missing bridging oxygen atom.

Moreover, another characteristic property of oxygen vacancies on TiO₂(110) was revealed: When a porphyrin derivative is laterally manipulated to the vicinity of an oxygen vacancy, the peak-positions of molecular orbital related resonances in dI/dV spectra shift to lower energies. This behavior is consistent with a positive charge localized at the vacancy sites.

Diffusion of Organic Molecules in Mesoporous Silica Materials

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The understanding of molecular dynamics in confined nanometer-sized geometries is one of the most important aspects in current research aimed to design novel smart materials based on host/guest systems such as drug-delivery materials. We investigate the translational, orientational and spectral diffusion of organic molecules in nanoporous silica materials using confocal and widefield fluorescence microscopy. The mesoporous channel system could be thought of a system of pipes in which molecules can diffuse. Analyzing the trajectories of the guest molecules by single particle tracking allows us to characterize the influence of the host system on molecular diffusion.

Two different studies are presented. First, we investigate simultaneously orientational-translational and orientational-spectral dynamics of single terrylendiimide (TDI) dye molecules as guests in the pores of an M41S host silica film synthesized with the ionic ammonium surfactant CTAB as template. Using confocal microscopy the in-plane angle of an individual molecule can be obtained with a precision of a few degrees, and can be correlated with the corresponding spectrum. Moreover, measuring simultaneously the position and the orientation of TDI shows molecules moving back and forth within the pore architecture. The movement is extremely linear, with the overall distance covered $\approx 1 \mu\text{m}$.

In a further step, short interfering RNA (siRNA) strands labelled with Atto 647N are incorporated into the pores of a thin M41S-film synthesized with the non-ionic Block-copolymer Brij-56 as template. RNA interference methods using siRNA represent a very promising strategy in the therapy of various different cancers. The diffusion of the biomolecules within the pore-system can be detected using widefield-microscopy. The understanding of the dynamics of the siRNA molecules in such mesoporous films is an essential prerequisite for their incorporation into novel silica drug-delivery systems.

Controlling DNA Devices with Chemical Oscillators and Switches

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The unique chemical and mechanical properties of DNA can be used to construct simple nanoscale actuators. Their principle of operation usually relies on the specific recognition between complementary DNA strands, the unzipping of DNA strands by DNA branch migration, and the fact that single-stranded DNA is a considerably more flexible molecule than DNA in its double-helical form. Based on these ingredients, reconfigurable supramolecular structures composed of flexible and rigid elements have been realized. With such structures, motion on the nanometer scale has been demonstrated as well as the binding and controlled release of other molecules. For future applications it will be desirable to make the action of DNA-based molecular machines dependent on environmental stimuli. Several methods are explored by which such environmental control can be achieved. We here show i) how DNA devices can be driven by oscillating chemical reactions, and ii) how DNA devices can be controlled using gene regulatory mechanisms.

[1] T. Liedl et al., *Nano Lett.* 5, 1894 (2005). [2] T. Liedl et al., *Angew. Chem. Int. Ed.* 45, 5007 (2006).

[3] W. U. Dittmer et al., *Nano Lett.* 4, 689 (2004). [4] W. U. Dittmer et al., *Small* 1, 709 (2005).

Programmable, Cytogenetic Submicroliter Lab-on-a-Chip: Detection of Chromosomal Imbalances

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Advances in molecular biology over the past decade have helped to enhance the understanding of the complex interplay between genetic, transcriptional and translational alterations in e.g. human cancers. These changes are the basis for an evolving field of high-throughput technique using microscopic amounts of patient-based tissue. On this account there is a high demand for the supply of DNA-microarrays as well as RNA- and Protein-microarrays. Currently different approaches aim to meet this increased request: SNP-arrays based on short (25mer) oligonucleotides, longer 60mer oligonucleotide arrays and BAC arrays with different density which cover the genome with a resolution of 1 Mb to 100 Kb. Tendency is the development of customized arrays with high variability in the diagnostic field. We present here a multifunctional *lab-on-a-chip* combining different platform elements like microdissection-, nanofluidic- and detection-modules. This chip combines serial processing with parallel downstream applications by using a minimum amount of genetic material as source for further investigations. Two different microdissection methods (atomic force microscopy and laser-based micromanipulation) provide the possibility to isolate samples in the range from several cells down to a single chromosomal band without the risk of contamination.

Instead of employing conventional micro-machined fluidics, we are using surface acoustic waves propagating on the surface of the substrate. This enables us to actuate smallest amounts of fluid (1 μ l down to 100 pl) along so-called "virtual tracks" without any channels or tubes. Such "virtual tracks" are fabricated by a monolayer surface treatment to laterally modulate the wettability of the substrate. The incompressibility of the fluid leads then to the streaming and continuous flow of the probe. Detection of the isolated, amplified and labelled genetic sample will be performed by plotting a 2-dimensional spot array onto the detection area of the chip and serves as a template for further array experiments.

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Balancing the White Light Shade of Dual-Color Emitting Nanocrystals

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It has been reported that solid state white light emitting devices would reduce the global electricity consumption by about 50%. Semiconductor nanocrystals (NCs) are highly fluorescent with fluorescence quantum efficiencies as high as 80%, being ideal candidates for use in white light emitting displays. Simple mixing of red, green and blue emitting NCs may lead to undesirable changes in color during the lifetime of the device due to different temporal stabilities of the components. Here we present the generation of white light from core-shell-shell CdSe/ZnS/CdSe NCs. These particles have two emission lines: blue (500 nm) from the CdSe shell and orange (612 nm) from the CdSe core. The ZnS layer is used as a spacer between the two emitting regions. The shade of the white light can be controlled during synthesis by varying the thickness of the CdSe shell. The relative intensities of both emission lines are also determined by intra-nanocrystal energy transfer. When the colors are well matched the resulting emission appears as pure white light. The synthesized NCs have 30% quantum efficiency.

Flow Equation Method for the Non-Equilibrium Anderson Impurity Model

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The Anderson impurity model is of central importance in correlated electron physics and is often used as a minimal model for studying quantum dots with Coulomb blockade effects. Steady-state non-equilibrium behavior can be obtained by applying a constant voltage bias across the impurity site. We examine this steady state by means of the flow equation method at zero temperature in the regime of weak to medium correlation strength.

In particular, we study the impurity orbital density of states and the decay of the quasi-particle resonance far away from equilibrium due to current-induced decoherence.

Programmable, Cytogenetic Submicroliter Lab-on-a-Chip: Detection of Genomic Imbalances in Solid Tumor Tissue

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Advances in molecular biology over the past decade have helped to enhance the understanding of the complex interplay between genetic, transcriptional and translational alterations in e.g. human cancers. There is a high demand for the supply of DNA-expression-microarrays. cDNA-arrays will provide valuable new insight into molecular identification and recognition of tumors as well as for diagnosis and prognosis of tumorigenesis. Currently indication-specific cDNA-arrays contain up to 7300 human gene

fragments. They are used for molecular sub-classification of tumors and for the identification of physiological processes which are known to be connected with tumorigenesis and -progression. Tendency is the development of customized arrays with high variability in the diagnostic field.

Here we present a multifunctional *lab-on-a-chip* combining different platform elements like microdissection-, nanofluidic- and detection-modules. This chip combines serial processing with parallel downstream applications by using a minimum amount of genetic material as source for further investigation. Laser-based microdissection provides the possibility to isolate samples in the range from several cells down to a single chromosomal band without the risk of contamination. Instead of employing conventional micro-machined fluidics, we are using surface acoustic waves propagating on the surface of the substrate. This enables us to actuate smallest amounts of fluid ($1 \mu\text{l}$ down to 100 pl) along so-called “virtual tracks” without any channels or tubes. Such “virtual tracks” are fabricated by a monolayer surface treatment to laterally modulate the wettability of the substrate. The incompressibility of the fluid leads then to the streaming and continuous flow of the probe. Detection of the isolated, amplified and labelled genetic sample will be performed by plotting a 2-dimensional spot array onto the detection area of the chip and serves as a template for further array experiments.

The use of this technique to interrogate biological information presents a paradigm change that has the potential to profoundly alter biology and medicine.

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A Simple Model System for Diffusion in Random Media

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Diffusion in heterogeneous, randomly structured media is a problem omnipresent in statistical and biological physics. An example from biophysics is the motion of proteins and stiff polymers in the cytoskeleton, the supporting frame of biological cells. Our model system is designed to mimic the essentials of this motion. It features a single point- or rod-like object, which moves through a fixed two-dimensional network of hard-core obstacles. Some very interesting features have already been found in these systems in case of ballistic motion [1]; we are now studying these models with underlying Brownian motion and a viscous fluid environment.

[1] F. Hoefling, T. Franosch, and E. Frey, unpublished.

Microencapsulated Ion-Sensors as a Novel Tool for in-situ pH Measurements

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Measuring analyte concentrations in small volumes is interesting for many applications. In cell biology, for example, the knowledge of local pH values has a great importance as pH changes are involved in many enzyme reactions and metabolic processes. Furthermore there is a significant need for the concurrent detection of various ions in good spatial and temporal resolution in vivo. Such an approach would help to understand the complex interrelationships between individual ion-concentrations and the role of each ion in mediating specific cellular responses. In diagnostics, concentrations of analytes have to be routinely measured in samples that are available only in small quantities. Usually the samples are lost after the measurement, it is complicated to measure several analytes in parallel, and also time resolved

measurements over extended periods of time are problematic (e.g. because of toxicity problems). Here we introduce polymer microcapsules with embedded ion-sensitive fluorescent dyes as local ion probes, which are potentially able to circumvent the above mentioned problems, as the fluorophore is retained in the picoliter-sized confinement of the capsule. In particular we report a new tool to measure the pH inside endocytotic vesicles in living cells. This innovative tool consists of a polyelectrolyte capsule fabricated by coating colloidal templates by using a layer-by-layer technique followed by dissolution of the core material. The analyte sensor is loaded inside the microcontainer. With this new technique several advantages can be achieved.

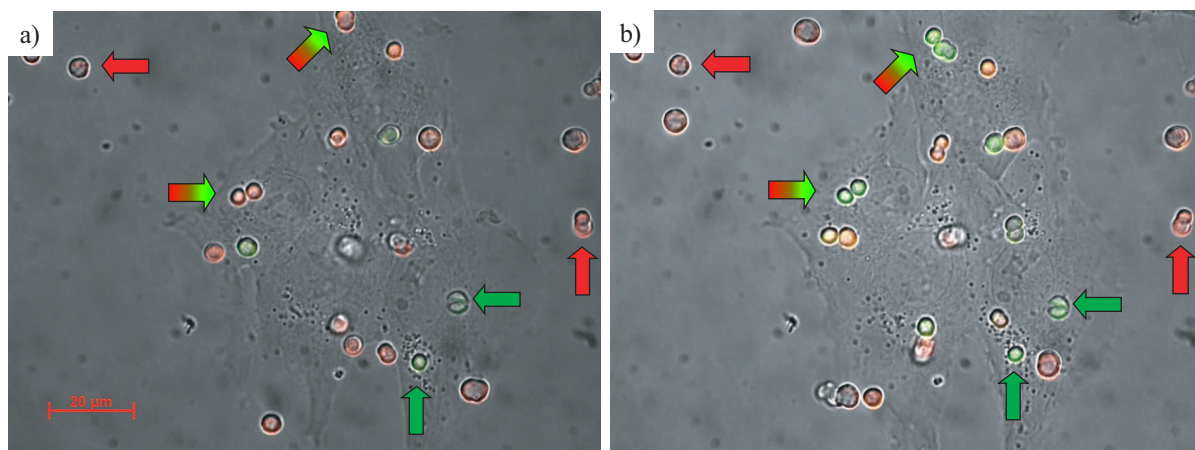


Fig.1 SNARF-1-dextran loaded microcapsules were added to the medium of MDA-MB435S breast cancer cells that are cultured on glass substrates. Image b) was recorded 30 minutes after. Image a) on the same cells. The fluorescence of the SNARF loaded capsules depends on their local surrounding pH value. Capsules that remain in the alkaline cell medium restrain their overall red fluorescence. Capsules that were already incorporated in the acidic endosome in the first image retain their green fluorescence. Some capsules were incorporated in endosomatic vesicles inside cells within the period of 30 minutes that had passed between the two images. Therefore these capsules are transferred from the alkaline environment outside the cells to an acidic environment inside the cells and thus change their fluorescence from red to green.

Construction of an Ultra Stable, High Resolution Optical Tweezer for Single Molecule Force Experiments on RNA Polymerase II

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Our goal is to gain insight into the behavior of single RNA Polymerase II (Pol II) enzymes at DNA lesions. Lesions, i.e. damages in DNA, occur frequently and have to be repaired in order to ensure the survival of an organism. One of the repair pathways is closely coupled to transcription, and is likely triggered by slowing down or stalling of the transcribing Pol II at the lesion site.

We are currently setting up an optical tweezer to monitor the process of transcription and transcription coupled repair in vitro. To obtain data with near base pair accuracy, the tweezer must show superior signal to noise ratio and low drift behavior. In order to meet this demand a special design is used.

Pseudo-Heterodyne Near-field Optical Microscopy

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Apertureless or scattering-type near field optical microscopy (s-SNOM) is a versatile optical imaging technique providing wavelength-independent resolution on the order of 10 nm. Its utility extends over the visible, infrared and microwave frequencies where it finds applications ranging from chemical and structural material identification to eigenfield mapping. Optical probing in s-SNOM is performed by a sharp, laser-illuminated probing tip scanned over the sample. Near-field interaction between the tip and the sample modifies the amplitude and phase of light scattered by the tip. The scattered light thus conveys the information on the optical properties of the sample. However, beside the useful signal generated by the near-field interaction, a large amount of direct scattering from the tip shaft is also produced. It represents the s-SNOM background signal and should be suppressed as much as possible since it can introduce various artifacts into s-SNOM images.

Here we present a new “pseudo-heterodyne” technique for s-SNOM signal detection [1] capable of background interference elimination in the entire near-UV to far-IR spectral range. It simultaneously measures near-field optical signal amplitude and phase by interferometric detection of scattered light utilizing a phase-modulated reference wave. By comparing the images obtained by the pseudo-heterodyne method to non-interferometric and homodyne interferometric detection methods, we show it provides reliable results even with weak nearfield optical contrasts, not reproduced correctly by the other two methods.

The pseudo-heterodyne detection thus represents the method of choice for s-SNOM investigation of systems yielding very fine optical contrasts. Its applications includes the detection of sub-10 nm Au particles [2] and spectroscopic identification of SiC polytypes [3], as well as biological objects such as single viruses [4].

[1] N. Ocelic et al., Appl. Phys. Lett. (accepted). [2] A. Cvitkovic et al., and R. Hillenbrand, Phys. Rev. Lett. 97, 060801 (2006). [3] A. Huber et al., Nano Lett. 6, 774 (2006). [4] M. Brehmet et al., Nano Lett. 6, 1307 (2006).

Efficient Photosensitization of C₆₀ Microcrystals with II-VI and III-V Semiconductor Nanocrystals

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Photoconductors are elementary and widely used components in optoelectronic applications such as photodetectors or photocopying machines. The sensitivity and spectral range of photoconductors can be drastically improved by sensitizers, which absorb light and provide additional charge carriers to the photoconducting material. At our poster we demonstrate that semiconductor nanocrystals (NCs) such as CdSe, CdTe and InP can efficiently photosensitize an array of needle-like C₆₀ microcrystals. A significant increase of the photocurrent (by 3 orders of magnitude) is observed for C₆₀ microcrystals covered with semiconductor NCs, in comparison with bare C₆₀ microcrystals. The photocurrent spectrum of the C₆₀ / NCs composites closely follows the NC absorption spectrum and can be tuned precisely by varying the NC size. The maximum external quantum efficiency was estimated by calculating the number of photoelectrons per incident photon and is about 10% for the CdSe / C₆₀ composites and about 3% for InP / C₆₀ composites. We explain the increased photoconductivity in the following way: The photoexcited electrons in the semiconductor NCs are transferred to the C₆₀ microcrystals causing photoconductivity, while the holes remain trapped in the semiconductor NCs.

Size Determination of (Bio-) Conjugated Water-Soluble Colloidal Nanoparticles – A Comparison of Different Techniques

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Gold-DNA conjugates were investigated in detail by a comprehensive gel electrophoresis study based on 1200 gels. A controlled number of single stranded DNA of different length was attached specifically via thiol-Au bonds to phosphine stabilized colloidal gold nanoparticles. Alternatively the surface of the gold particles was saturated with single stranded DNA of different length either specifically via thiol-Au bonds or by nonspecific adsorption. From the experimentally determined electrophoretic mobilities estimates for the effective diameters of the gold-DNA conjugates were derived by applying two different data treatment approaches. The first method is based on a making a calibration curve for the relation between effective diameters and mobilities with gold nanoparticles of known diameter. The second method is based on Ferguson analysis which uses gold nanoparticles of know diameter as reference data bas. The obtained results help to understand the binding of DNA to gold nanoparticles. In particular the specific attachment of DNA via gold-thiol bonds to Au nanoparticles is compared to nonspecific adsorption of DNA. Also the maximum number of DNA molecules that can be bound per particle was determined. In order to generalize the investigation also more complex particle systems and other techniques were used. In particular gold and CdSe/ZnS particles embedded in a polymer shell with attached PEG were investigated. The size of these conjugates was measured with different techniques including transmission electron microscopy (TEM), gel electrophoresis, high performance liquid chromatography (HPLC), light scattering, fluorescence correlation spectroscopy (FCS), and thermophoresis. The results are critically compared and advantages and disadvantages of the respective methods are discussed.

Nanoparticle Cellular Uptake Mechanism

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The structure and properties of nanoparticles make them useful for many biological applications. Despite of that, little is known about the mechanisms which lead to their incorporation into living cells. We want to determine the effect of size, shape and surface modification on the bioavailability, uptake rates and subcellular distribution of nanoparticles. Transmission electron microscopy and the high contrast of colloidal gold nanoparticles allow us to trace them inside cells. We present and discuss first images.

SAW-induced Quantized Current Transport in Carbon Nanotubes

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Carbon nanotubes (CNT) are an example for natural one-dimensional conductors and therefore interesting objects for low dimensional electronics. Electrical transport through CNT is investigated since their discovery and also the possibility of forming quantum dots in a contacted CNT.

We present an alternative way of current transport through such a quantum dot by applying surface acoustic waves (SAW). Dissolved CNT were aligned on a prestructured LiNbO₃-substrate and after drying current transport measurements have been carried out to show the coulomb blockade behaviour of the CNT. If a SAW is launched with a wavelength double the size of the electrode distance ($f \approx 1$ GHz), the attendant electric potential of the wave modulates the source and drain barriers of the quantum dot periodically. While lowering the source barrier an electron can tunnel into the dot, half a wavelength later the drain barrier is lowered and the electron can turn out again.

Thus a very precise quantized current ($I = e \cdot f$, e : elementary charge, f : frequency of the SAW) can be created by transporting one electron per wave period through the quantum dot. This current quantization can be shown in a stepwise increase of current in dependence of the SAW-amplitude.

Intracellular Transport meets Spintronics

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Traffic phenomena occur in biological contexts as well as in mesoscopic quantum systems. Molecular motors move along parallel one-dimensional filaments in cells, serving as biological transport engines. On the other hand, spintronics devices aim to exploit quantum effects, the spin of electrons, when passing these through nanowires. Here, we present a generic model that underlies both situations. Allowing particles in an exclusion process to possess internal states, the latter account for several parallel lanes as well as for different spin states, where Pauli's exclusion principle is respected. Exploring the system's behavior, we find that it can be tuned by controlling the particle fluxes at the boundaries. In particular, a spontaneous polarization may occur at a certain spatial position and, upon changing the fluxes at the boundaries, be driven in or out of the system. We derive the shape of the density profiles as well as resulting phase diagrams analytically by a mean-field approximation and a continuum limit. Continuous as well as discontinuous lines of phase transition emerge, their intersections induce multicritical behavior. Domain wall theory allows us to analytically study the delocalization of a domain wall when approaching a delocalization transition.

Single-Molecule Mechanics of Actin-Crosslinking Proteins

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While the mechanics of actin-crosslinking molecules itself is quite well characterized using single molecule force spectroscopy, the interplay between cross-linking molecules and actin-filaments is mainly studied in bulk-experiments. Data on the interaction between single cross-linking proteins and actin-filaments is almost entirely missing. We developed two multiple optical tweezers assays which will provide insight in the inter-molecular mechanics of the cytoskeleton on the level of single molecules. Two actin-filaments which are decorated with a cross-linking protein are moved relatively to each other. Thus transient binding and unbinding between the two filaments can be induced. These experiments provide insight in the force-dependent kinetics of the actin – crosslinker – actin system and will answer e.g. the longstanding question, if the unfolding of single domains of cross-linker molecules might be of physiological relevance.

Simulation of DNA Based Nanostructures in Nanotechnology

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Structural DNA technology is a new and promising field, based on the common biochemical notion of structural complementarity between the two strands of a DNA double helix. It exploits the Watson-Crick base pairing of DNA to create stable branch-points with sequence control, resulting in highly predictable structures in two- and three-dimensions. Therefore DNA double helix structures are emerging as a useful scaffold for creating nanostructures and as components for nanomechanical devices [1].

In this work we use theoretical approaches for the study of three-arm junction structures, that are comprised of three double-helical arms that are connected in a branch-point to form a Y-shaped structure [2]. Our objective involves the study using molecular modeling of a single unit and possible nanostructures that can be formed using the three-arm junction as building blocks. This method can be used to interpret and guide experiments and synthesis of new nanostructures.

Two three-arm junction structures were constructed based on DNA sequences defined by the Biomolecular Nanoscience Group using the nucleic acid builder program Namot2 version 2.2 [3]. An established

protocol [4] was adopted for the dynamic simulation of both three-dimensional models. All simulations have been carried out using the NAMD suite of programs [5] and the AMBER-parm-98 force field [6]. After 5 nanoseconds simulation we made a detailed analysis of both structures taking into account stability, flexibility, deformability and H-bonds formation. We observe some features that help us to interpret experimental results obtained by the Biomolecular Nanoscience Group.

Support: PROBRAL (CAPES/DAAD) and ENNAB

[1] N. C. Seeman, Nature 421, 427 (2003). [2] F. J. J. Overmars et al., J. Mol. Biol. 255, 702 (1996). [3] C. S. Tung et al., Comput Appl Biosci. 10, 427 (1994). [4] D. L. Beveridge et al., Biopolymers 73, 380 (2004). [5] L. Kalé et al., J. of Comp. Phys. 151, 283 (1999). [6] W. D. Cornell et al., J. Am. Chem. Soc. 117, 5179 (1995).

Towards Artificial Viruses for Cancer Gene Therapy: Novel Biodegradable Gene Carriers Based on Pseudodendritic Oligoamines

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Biodegradable gene carriers with a pseudodendritic core structure were generated from low molecular weight oligoethylenimine 800 Da. Core characteristics and surface modifications strongly influenced biophysical and biological properties and out of them we were able to draw structure - response relationships. Increasing chain length of oligoamines on the surface resulted in enhanced DNA-binding ability while cytotoxicity and transfection efficiency of pseudodendrimers were affected of both core characteristics and surface modification.

Polycationic polymers have been continuously improved for gene transfer. Since the standard cationic polymer polyethyleneimine (PEI) is known to be highly toxic, there is the urgent need to develop non-toxic and biocompatible polycations for safe and efficient gene delivery. Low molecular weight (LMW) polyamines like branched oligoethyleneimine 800 Da (OEI) show low toxicity but are not able to mediate gene transfer efficiently due to a lack of DNA-complex stability in physiological environment [1,2]. To enhance transfection efficiency pseudodendritic conjugates based on OEI were formed.

OEI represents the centre of these pseudodendrimers and will mediate endosomal release of pseudodendrimer/DNA – complexes by the “proton sponge effect”. By coupling an excess of different dioldiacrylates to OEI pseudodendritic cores were formed. All core branches contain two labile ester bonds which can break down at low pH in intracellular medium and should be responsible for the biodegradability of the polymers. Varying spacer length in between the esters and different surface modifications with oligoamines upon the pseudodendritic core should influence conjugate parameters such as charge density, degree of branching and molecular weight which play an important role in DNA-binding, particle structure and transfection efficiency.

[1] J. Kloeckner et al., Bioconj. Chem., in press. [2] J. Kloeckner et al., Europ. J. of Pharmac. Sci., in press.

Observation of Nanoparticle Uptake in Living Cells by Single Particle Tracking

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Nanoparticles consisting of DNA complexed by cationic polymers (polyplexes) can be used as non-viral vectors for gene transfer into cells and are an important candidate for gene therapy. However, they are still less efficient than viral vectors. One reason is the lacking cell specificity of non-viral vectors

compared to viral vectors. To enhance cell targeting, PEI (polyethyleneimine) polyplexes with a PEG (polyethyleneglycol) shield were functionalized with EGF (epidermal growth factor) for specific binding to the EGF receptor on the cell surface. To analyze uptake dynamics and kinetics of PEI, PEI-PEG and PEI-PEG-EGF polyplexes at a single particle level, we transfected HUH7 cells with these particles. By using highly sensitive fluorescence wide-field microscopy, single particle tracking was performed to generate trajectories of the uptake dynamics. The extracellular particles were quenched at various times to determine the relative amount of internalized particles. Typically, three types of particle motion were observed: (1) a phase of immobility and slight drift with very small step lengths that could be assigned to membrane movement; (2) a diffusive movement characterized by a larger step length that was independent of membrane movement; and (3) directed motion along microtubules. No diffusive or directed motion was observed before particles entered the cell. This suggests that internalization took place in the first immobile phase. PEI-PEG-EGF particles are internalized up to 100% within 10-15 minutes whereas PEI and PEI-PEG particles show internalization to a much lesser extent.

NRG Matrix Product States and Quantum Information Processing

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We use newly developed “Matrix Product” techniques to compare the Numerical Renormalization Group approach for the Single Impurity Anderson model to the Density Matrix Renormalization Group approach. We would also discuss the perspectives for applying Matrix Product techniques to time-dependent models relevant for the field of quantum information theory, such as a qubit coupled to a bath of decohering excitations.

AFM as a Tool to Probe the Mechanics and Kinetics of Integrins on Living Cells

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Lymphocyte recruitment from the bloodstream plays an important role in a variety of physiological and pathological processes. Intercellular adhesion mediated by the integrin VLA-4 and its endothelial ligand VCAM-1 precedes the extravasation of the lymphocytes to the target tissue. Integrins participate in all adhesive steps between tethering and rolling and firm adhesion. To match this bridging position, these cell surface receptors are subject to insight-out and outside-in signaling, by which the kinetics and mechanics of adhesion under the shear force in the blood stream is regulated.

Here, atomic force microscopy experiments are employed to investigate the attachment of VLA-4 bearing cells to VCAM-1 coated surfaces. We introduce a combined interpretation of classical force spectroscopic parameters, such as forces and rupture lengths, with a new analysis of curve shape and bond formation rate. Based on the assumption that the force-distance curves describe the extrusion of membrane tethers, our interpretation of the data gives insight into a multitude of kinetic and nano-rheological parameters. We show that AFM can be used to simultaneously measure the physiological off-rate under force and the two-dimensional on-rate of proteins immobilized on different surfaces, as well as nano-viscoelastic parameters of the receptor anchoring membrane. We show that the careful analysis of these parameters in combination with flow chamber experiments allows us to fully describe the effect of an VLA-4 activating agent, Mg²⁺, in living cells.

Force Spectroscopy on Single Siloxane Polymers

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Siliconelastomers are high-performance materials, used in a wide field of applications. Among their remarkable qualities are low temperature dependence of the mechanoelastic properties, high resilience against thermooxidation, high flexibility at low temperatures, adhesive behavior and physiological harmlessness. On the other hand siliconelastomers also have some disadvantages like low ultimate mechanical strength, vulnerability to hydrolytic reagents and moisture expansion in unpolar environment.

Single molecule force spectroscopy using an AFM offer a unique possibility to investigate the mechanical performance of single siloxane polymer chains.

Phase-Space Theory for Nonlinear Detectors of Superconducting Qubits

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Circuit quantum electrodynamics schemes offer an attractive new tool to harness the interaction between coherent matter systems and the electromagnetic field similar to quantum optics. We study the decoherence of a quantum bit (qubit) due to the coupling to a damped harmonic oscillator. The state of the qubit is typically read out by a dispersive measurement of the cavity. We consistently model the backaction in this system using a phase-space approach and show that the qubit dephasing rate can be proportional to the photon decay rate of the cavity κ or to $1/\kappa$. We explain these results in terms of a crossover from a novel strong coupling regime to weak coupling explained in terms of a phase Purcell effect.

Fabrication of Nanoparticle Chains with Rolling Circle Amplification

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The well-established biochemical technique of rolling circle amplification allows for a simple fabrication of long single-stranded DNA molecules with a repeating sequence motif. In RCA, small circular single-stranded oligonucleotides act as templates for the polymerization of the complementary strand. A DNA polymerase with an efficient strand displacement activity – such as the polymerase from phage phi29 – can processively copy the circular template without stopping. This results in a long DNA strand with periodic sequence. We here demonstrate that this method – using DNA recognition and biotin-streptavidin binding – provides a simple procedure for DNA-directed nanoscale organization of nanoparticles. As an example, a 74 nucleotide long DNA molecule is amplified into a sequence-periodic single strand with a length up to several micrometers. Hybridization of this long periodic DNA template to the biotinylated complement of the sequence motif results in a long DNA duplex with a periodic arrangement of biotin binding sites. We show that this template can be used, for instance, to arrange streptavidin coated gold nanoparticles.

[1] S. Beyer et al., Nano Lett., 5, 719 (2005).

Time Resolved Energy Transfer from Single Colloidal Semiconductor Nanorods to Single Dye Molecules

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We investigate nanoscale energy transfer between single CdSe(CdS) core-shell nanoparticles and single dye molecules using the time correlated single photon counting (TCSPC) technique. Such a hybrid system of nanoparticles and molecules bears several advantages: the large absorption cross-section and exceptional photostability of inorganic semiconductors can be combined with the large luminescence quantum yield of dyes resulting in a high overall photoluminescence efficiency due to energy transfer. By appropriate selection of the optical absorption of nanoparticles and dye molecules we ensure exclusive excitation of nanoparticles. On the other hand, spectral overlap of the dye's absorption with the photoluminescence of the nanocrystals makes energy transfer possible. Time resolved measurements on single nanocrystals clearly show a decrease of the nanocrystal's luminescence lifetime in the presence of dye molecules indicating energy transfer. This quenching of the donor fluorescence strongly depends on the concentration of dye molecules in the film, which controls the average distance between nanoparticles and dye molecules. When illuminating the sample under our wide-field imaging microscope we observe a local increase of the dye's photoluminescence at the spatial positions of the individual nanoparticles. We now hope to identify the temporal rise in the emission from a single dye molecule due to energy transfer from the nanocrystal. Such a rise in emission was recently reported in the spectroscopy of single molecules of dye end-capped conjugated polymer molecules, which act as covalently bound donor-acceptor systems [1].

In conclusion we are able to demonstrate energy transfer from nanoparticles to surrounding dye molecules on a nanometer scale.

[1] K. Becker et al., J. Am. Chem. Soc. 128, 6468 (2006).

Decorated Nanowires, Nanorings and "Hedgehog-Like" Superstructures Containing CdSe Nanocrystals, Produced by Microwave-Assisted Synthesis

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Semiconductor nanocrystals prepared in water solution have a number of possible applications ranging from light emitting diodes to biological labels. Herein we studied microwave assistance synthesis of CdSe quantum dots. Cadmium perchlorate and N,N-dimethylselenurea were used as sources of Cd and Se ions. Different stabilizers: sodium citrate, glycine, and different reaction condition (pH and reaction times) were used. Quantum efficiency of nanocrystals formed was not high (less than 3%), but by varying the reaction conditions it is possible to play with particle morphology. For solutions with citrate as stabilizer mostly spherical particles grow, but "hedgehog-like" superstructures can also be made. The use of glycine and glycoic acid leads to nanowires, and nanorings decorated by spherical nanoparticles. It is also possible to make core-shell structures, which slightly increases quantum efficiency.

Spectroscopic Nearfield Microscopy Powered by THz Frequency Combs

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Recently we reported the application of a new type of Fourier transform spectroscopy called coherent frequency-comb FTIR [1]. This multi heterodyn detection principle, allows a fast and parallel readout of infrared spectra without any moving parts in the setup.

Now this spectrometer is being modified to employ Terahertz (THz) waves, utilizing a commercial large area THz emitter and electro-optic detection.

This new spectrometer will then be used to illuminate and read out a scanning near-field microscope (SNOM). This way we overcome the diffraction-limited resolution of THz waves, (1 THz corresponds to a wavelength of 300 μm !) and expect resolutions of about 30 nm. The near-field microscope is based on an AFM with commercial, cantilevered tips and works on the principle of light scattering; thus it is named scattering or s-SNOM.

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Nanosystems From the Viewpoint of Quantum Impurity Models

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One of the central themes of quantum mechanics is to understand how the dynamics of a quantum system is influenced by its environment. A specific, prototypical class of models that embody this theme and arise in a broad range of problems in condensed matter physics are quantum impurity models (in modern parlance, they might as well be called qubit models): a quantum degree of freedom with a small number of discrete states, the “impurity” (or qubit), is coupled to a continuous bath of excitations. The environment can arise from a fermionic conduction band, such as in the Anderson impurity model, currently relevant for describing quantum dots coupled to leads or for the treatment of correlated lattice models using dynamical mean field theory; or it can be of bosonic character, as in the spin-boson model, presently popular for modelling the decoherence of qubits. As the solution to these systems can be enormously complex and computationally challenging in the limit of strong coupling, we present a straightforward and intuitive approach based on matrix product state as a hybrid method between numerical renormalization group (NRG) and density matrix renormalization group (DMRG).

Surface Treatment of SU-8 and Characterisation

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Micrometer structures for fluidic systems are often manufactured in polymers such as SU-8 or PDMS. Unfortunately both polymers are highly hydrophobic. With the decrease in dimension, the role of capillary forces becomes important. Thus, controlling the surface free energy (γ_s) and the wetting behaviour of the system is crucial for microfluidics [1]. Commonly, a dry etch treatment by an O₂ plasma process is utilized to render polymeric surfaces hydrophilic [2]. Alternatively, wet chemical etching procedures can also alter the surface contact angle [3]. Both techniques increase the surface roughness and change the surface chemistry.

The development of γ_s with respect to time due to ageing effects has been monitored over three months (Fig. 1) to calculate the capillarity and to estimate the shelf-lifetime of bio-MEMS devices. To gain a better understanding of the time-dependent hydrophobic recovery we carried out contact angle goniometry and calculated γ_s from this data. Topographic imaging by atomic force microscopy (AFM) allowed

the quantification of changes of the surface roughness induced by the etching processes. In order to correlate our results with surface chemical composition, XPS measurements were performed.

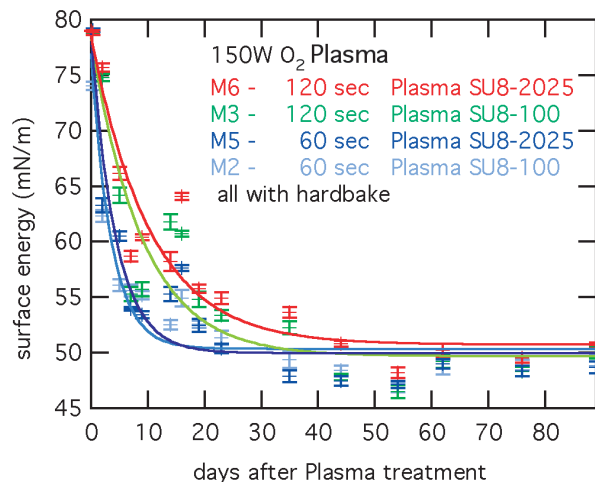


Fig.1 The development of γ_s has been monitored over three month

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Light Driven Microfluidic

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Microfluidics is a quantum leap for complex liquid manipulation in a wide variety of biological and chemical applications of the life sciences. Liquid is pumped and switched through lithographically defined channels on the micron scale. However, microfluidic chips connect to a considerable complex interface overhead for pumping, switching and providing the liquids.

Here we present an all-optical fluid flow control, which allows highly flexible and dynamic liquid handling, both in microfluidic channels and in two-dimensional gels. The fluid follows the arbitrary shaped two-dimensional path of an infrared laser scanning microscope without the need for microfluidic tubings. The flow is driven by temperature dependent viscosity under fast thermal expansion.

Pump speed rises quadratically for decreasing thickness, reaching 20 $\mu\text{m/s}$ in 2.5 μm thin water films. Liquids of high viscosity are equally pumped. The technique has the potential to free micro- and nano-fluidics from channels, valves, delicate interfacing and complex pump design.

Roughening of 2D Nanoparticles by Three-Body Interactions

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In the majority of cases adsorbed particles on solid surfaces form two-dimensional, nanometer-sized islands at some coverage. Island formation is important for all processes that depend on the distribution of adsorbates and require surface diffusion, such as in heterogeneous catalysis. An unclear aspect is the shapes of the islands. Using high-speed scanning tunneling microscopy (STM) we have investigated fluctuating islands of adsorbed O atoms on Ru(0001). The data show surprisingly ramified islands pointing to complex interactions between the adsorbed O atoms. Density functional theory (DFT) calculations at

coverages as low as 1/24 of a monolayer reveal attractive pair-wise interactions between the O atoms that can explain the (2×2) structure of the islands. However, the calculations also find repulsive three-body interactions between the adsorbed O atoms. Monte-Carlo (MC) simulations including such three-body interactions quantitatively reproduce the experimentally observed atom distributions. That many-body interactions are important for adsorbate systems has been recognized previously. They have been made responsible for asymmetries in phase diagrams of adsorbate layers, and have been used to explain the shapes of small clusters of adsorbed atoms, in particular of metals. The data presented here go beyond these investigations, as the influence of many-body interactions on the ordering behavior of adsorbed particles could be monitored with atomic resolution and in real time for a system in thermodynamic equilibrium. The repulsive three-body interactions are significant – they are of similar magnitude as the attractive pair-wise interactions – and hence are necessary for a complete understanding of the energetics of adsorption, even at the low coverages of applied surface processes such as catalysis .

Large Scale Local Probe Oxidation of n-Octadecyltrichlorosilane Monolayers with High Resolution

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Scanning probe patterning techniques such as dip-pen nanolithography (DPN) and oxidative scanning probe lithography have been demonstrated to be very promising methods for the fabrication of functional nanodevices [1,2]. Using self-assembly techniques the created patterns can be used as templates for local chemical modification, introducing functionality on spatially defined spots on a substrate. Reports on the fabrication of nanoscale electrical circuits, multi-component nanoparticle and protein arrays have been published recently. In this publication we demonstrate the automation and large scale surface patterning using an automated scanning probe microscopy setup for the local probe oxidation of octadecyl trichlorosilane (OTS) monolayers on silicon substrates. The electro-oxidation of OTS monolayers has been demonstrated to be a versatile robust platform for a broad range of surface modification reactions. During the oxidation process the terminal methyl groups of the OTS monolayer are converted into carboxylic acids (-COOH) groups [3]. Subsequently the formed patterns of carboxylic acid groups can be used as a template in additional surface decoration steps. The application of high-resolution, large scale surface patterning could be of great importance for applications such as sensors. Additionally patterning of large areas enables the application of surface characterization techniques such as XPS or IR.

Using a tailor-made automated AFM with dedicated software and a programmable sample stage we demonstrate the automated patterning of large areas on a surface with high resolution. Although the final results depend on tip quality we have demonstrated the writing and subsequent surface functionalization of a large number (>1000) of patterns on a millimeter-sized substrates using a single AFM-tip covering the gap from nanometer patterning to millimeter-sized applications. The surface oxidation may be performed in two distinct modes. In the first mode the tip is rastered over the sample surface while either continuously applying a bias voltage or selectively applying bias voltages using an imported gray-scale image as template. In the second mode the tip follows a pre-defined vector trajectory over the surface. Because this is performed without the need of pre-imaging the surface tip-life is greatly enhanced.

The obtainable resolution depends critically on the diameter of the used tip and may be improved lines with a width down to 12 nm (FWHM) by using uncoated highly doped Si-tips at higher bias voltages (up to 50 V) instead of metal coated cantilevers. In addition, we will present the first steps towards large scale surface patterning by using arrays of, initially, 4 parallel cantilevers.

[1] D. S. Ginger et al., *Angew. Chem.* 116, 30 (2003); *Angew. Chem. Int. Ed. Engl.* 43, 30 (2003). [2] D. Wouters et al., *Angew. Chem.* 116, 2534 (2004); *Angew. Chem. Int. Ed. Engl.* 43, 2480 (2004). [3] a) R. Maoz et al., *Adv. Mat.* 12, 725 (2000); b) D. Wouters et al., *Langmuir* 19, 9033 (2003); c) S. Hoepfener et al., *Adv. Mat.* 14, 1036 (2002).

Investigation of Silicon Cantilevers for Bio-Application

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The atomic force microscopy (AFM) has been proved to be quite a useful tool in nanoscience since its invention in the 1980s. It is not only powerful in imaging but also very potent in small mass/force detection, atomic manipulation, etc. Of these areas, the bio-application attracts much attention nowadays, including single DNA detection, living cell real time imaging, viscoelasticity characterization and so on. Since in most of these experiments, the AFM cantilever is operated in liquid environment and then the damping of the cantilever becomes a major problem which limits the accuracy and resolution of the measurement. So how to improve the quality factor of these AFM cantilevers is the main part of my research. Many different types of silicon cantilevers are fabricated in the nanotool facility of McGill University, and the frequency and Q of these cantilevers are measured.

Templated Synthesis of Metal-DNA Junctions Using Phenanthroline-DNA Conjugated Molecules

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Metal coordinated DNA molecules are important nanobuilding blocks due to their well-defined geometries, redox and luminescent properties. We have synthesized novel phenanthroline-DNA conjugated molecules for use as nano-building blocks. Copper(I), copper(II) and silver(I) metal-DNA junctions were synthesized using external or internal templated DNA strands. The properties of these metal-DNA junctions were then investigated through thermal denaturation, circular dichroism and fluorescence spectroscopy. Further study of these junctions for the construction of novel DNA nanostructures is currently underway.

Hybrid Systems of Single Wall Carbon Nanotubes and Nanocrystals: Synthesis and Optoelectronic Charge Transport Properties

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Colloidal II-VI semiconductor nanocrystals (NCs) have fascinating optical and optoelectronic properties, and therefore they are widely used in biolabeling applications. In this project, individual colloidal nanoparticles are contacted via single wall carbon nanotubes (SWNTs) as mesoscopic contacts. To this end, the self-assembly of hybrid systems consisting of single wall carbon nanotubes and either colloidal gold or II-VI semiconductor nanocrystals like CdTe is presented. To form such a system of SWNTs and NCs one of the strongest non-covalent and irreversible bindings in biology, the biotin-streptavidin van-der-Waals interaction, is used [1]. The SWNTs bound to a nanocrystal or several ones are then contacted by methods of electron beam lithography. We characterize the physical properties of such a mesoscopic circuit by means of optoelectronic charge transport measurements. The optoelectronic low temperature measurements of quantum transport through such a mesoscopic device involve the application of a source-drain voltage with an additional gate potential to observe single electron effects like Coulomb-blockade [1].

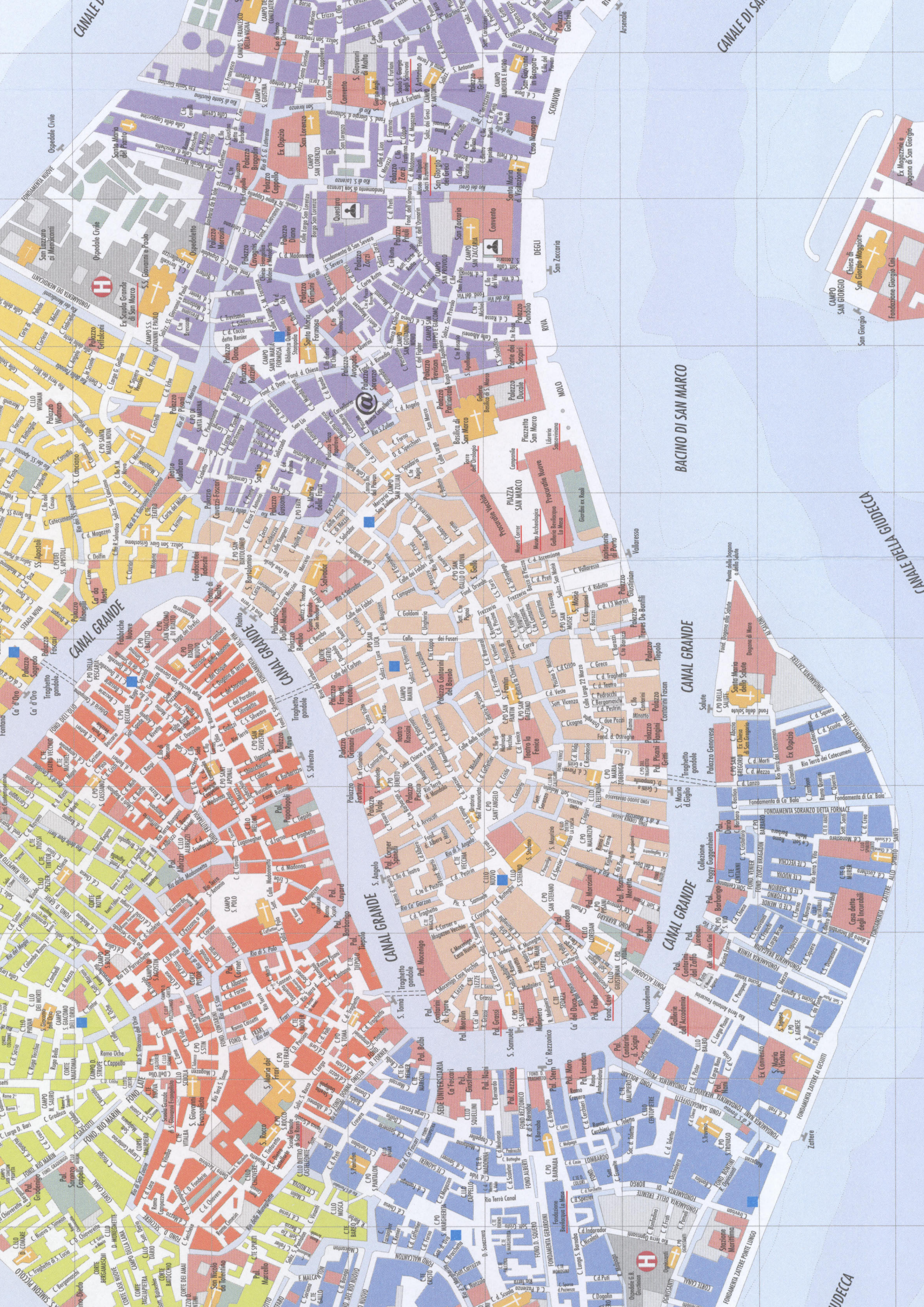
[1] T. Smorodin et al., Nanotechnology 16, 1123 (2005).

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Workshop Location

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Conference office (during conference): +39-041-2719 550

Hotel List

- ISTITUTO CANOSSIANO
Fondamenta de le Romite, 1323 Dorsoduro, 30123 Venezia
Tel: +39 041 240 9711, Fax: +39 041 240 9712, Email: cvenezia@fdcc.org
Close to the Accademia brigde (Ponte Accademia).
The house closes at midnight unless other arrangements are made.
- DOMUS CILIOTA
Calle delle Muneghe, S. Marco 2976, 30124 Venezia
Tel: +39 041 520 4888, Fax: +39 041 521 2730, Email: info@ciliota.it
The hotel is located in the historic center of Venice.
Ask for the "Calle delle Muneghe" in the St. Mark's quarter (Sestiere San Marco).
- RESIDENZA GOLDONI
Calle della Fava or Stagneri, San Marco 5234, 30124 Venice
Tel: +39 041 241 0086, Fax: +39 041 277 4728, Email: info@residenzagoldoni.com
Close to the Rialto bridge.
- CASA SANTA MARIA FORMOSA
Castello 5841, 30100 Venice
Tel: +39 041 241 1300, Fax: +39 041 241 5982, Email: info@casiformosa.com
Close to the church Santa Maria Formosa.
- LOCANDA LA CORTE
Castello 6317, 30122 Venice
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Close to the church Giovanni e Paolo.
- RESIDENZA CA' SAN MARCO
Calle de le Balote
San marco 4868/4869 San Marco
Tel: +39 041 241 3864, Fax: +39 041 241 7245, Email: info@hotelcasasanmarco.it
Close to the Rialto bridge.
- SANTA MARIA DELLA PIETA
Castello - Calle della pietà 3701, 30122- Venezia
Tel: +39 041 244 3639, Fax: +39 041 241 1561, Email: info@pietavenezia.org
Next to the boat landing "San Zaccaria" for San Servolo.
- FORESTERIA VALDESE DI VENEZIA
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Notes

Notes

Notes

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Tuesday, 26.09.

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Erdmann	Matthias
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Huth	Florian
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Schmitz	Julia
Schwaderer	Peter
Serban	Ioana
Sobey	Tom
von Ribbeck	Hans-Georg
Weichselbaum	Andreas
Weinert	Franz
Wouters	Daan
Zebli	Bernd

Postersession II

Thursday, 28.09.

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Wintterlin	Joost
Xuefeng	Duan
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CeNS Workshop: "Emerging Nanosystems - From Quantum Manipulation to Nanobiomachines", 25.09. - 29.09.2006, Venice International University

Time	Monday 25. Sep 06	Tuesday 26. Sep 06	Wednesday 27. Sep 06	Thursday 28. Sep 06	Friday 29. Sep 06
9:30	Opening	-	-	-	-
9:45	Paul Hansma High speed atomic force microscopy and the molecular mechanics of bone fracture	Hermann E. Gaub Torturing molecules by AFM	Theodor W. Hänsch Towards a quantum laboratory on a chip	Ulrich B. Wiesner Nanohybrids and nanobiohybrids	Irmgard Frank Chemical reactions and dynamics in nanosystems: First-principles simulations
10:30	Thomas Basché Electronic excitation energy transfer between two single molecules	Kevin Hennessy Coupled excitons and photons in photonic nanocavities	David Bensimon Single molecule studies of chromatin remodelling factors	Jens Ebbecke Acoustic waves in semiconductor nanostructures	Eli Yablonovitch The impedance-matching predicament: A hurdle in the race toward nano-electronics
11:15	<i>Coffee break</i>	<i>Coffee break</i>	<i>Coffee break</i>	<i>Coffee break</i>	<i>Coffee break</i>
11:45	Fritz Keilmann Spectroscopic near-field microscopy with infrared frequency-combs	Thorsten Hugel Scrutinizing nanobiomachines with single-molecule fluorescence and force spectroscopy	Piet Brouwer Current-induced magnetization dynamics in nanoscale magnets	Stephanie Hoeppeener Nanolithography and self-assembled monolayers - Platforms to assemble functional structures	Jochen Feldmann Nanocrystals for photonic applications: towards synthetic light harvesting complexes
12:30	<i>Lunch</i>	<i>Lunch</i>	<i>Lunch</i>	<i>Lunch</i>	<i>Lunch</i>
14:30	Gerhard Abstreiter Manipulating and controlling semiconductor quantum dots	Enrico Gratton Particle tracking in 3D: Chromatin structure and dynamics	Informal discussions		
15:15	Erik Winfree Algorithmic self-assembly of DNA	Harold G. Craighead Nanostructures for biomolecular analysis	Thomas W. Ebbesen Surface plasmon photonics		
16:00	<i>Coffee break</i>	<i>Coffee break</i>	<i>Coffee break</i>		
16:30	William M. Shih DNA-based molecular containers	Thomas Franosch Phase separation in driven non-equilibrium transport	Vahid Sandoghdar On the coupling of a single emitter to photons		
17:15	Don C. Lamb Single molecule investigations: From protein dynamics to virus entry	Christoph F. Schmidt Eg5: a four-headed mitotic kinesin motor	Andreas R. Bausch Physics of complex actin networks: from molecules to networks		
18:00	Welcome reception				
19:00	<i>Dinner</i>		<i>Dinner</i>		
	Poster Session I		Poster Session II		
	Departure				