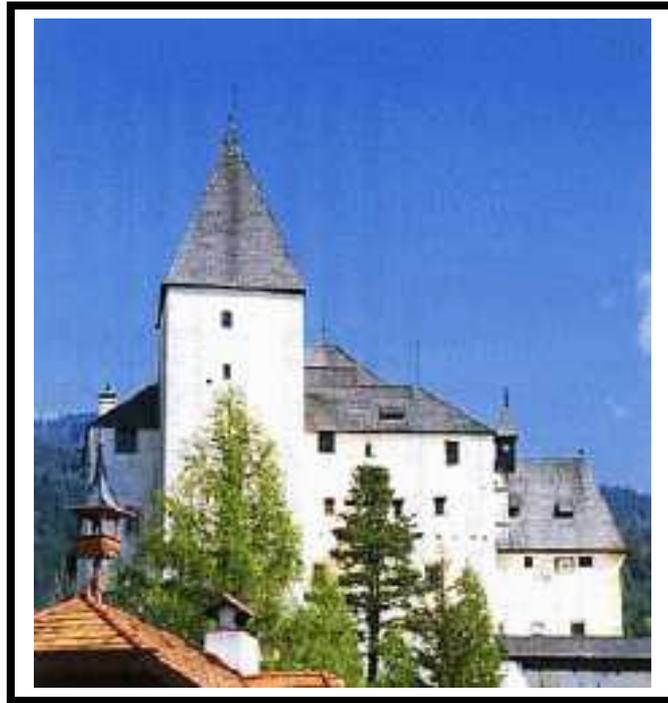


CeNS Winterschool 2001



Sensing and Manipulating in the Nanoworld

Mauterndorf, Austria

18 - 23 February 2001



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**Sensing and Manipulating
in the Nanoworld**

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Sponsored by:

Deutsche Forschungsgemeinschaft via
SFB 486: „Manipulation von Materie auf der Nanometerskala“
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SFB 513: „Nanostrukturen an Grenzflächen und Oberflächen“

BMBF via a Max Planck Research Award

Kompetenzzentrum Nanoanalytik

Program

Sunday, February 18th

17.00 – 19.00 Registration

Monday, February 19th

8.30 – 8.45 Opening

8.45 – 9.45 **V. Colvin** (Rice University):
Nanochemistry and Colloids: The Basics

9.45 – 10.45 **J. F. Stoddart** (Department of Chemistry and Biochemistry, University of California):
Artificial Molecular Machines

10.45 – 11.15 Coffee / Tea

11.15 – 12.15 J.-M. Caruge, **M. Orrit** (C.P.M.O.H., CNRS et Université Bordeaux I):
Single molecule spectroscopy : probing of charge transport at a nanometre scales

12.30 – 17.00 Lunch and informal discussion

17.00 – 17.15 Coffee / Tea

17.15 – 18.15 **H. Craighead** (Applied Physics, Cornell University):
Nanomechanical Systems

18.15 – 19.15 **J. Dhont** (Forschungszentrum Jülich):
Spinodal Decomposition of Colloids

Tuesday, February 20th

- 8.45 – 9.45 **B.N.J. Persson** (FZ Jülich):
Nanomechanics, adhesion and friction
- 9.45 – 10.45 **T. Basché** (Institut für Physikalische Chemie, Johannes Gutenberg-Universität Mainz):
Microscopy and Spectroscopy of Single Nanoparticles I
- 10.45 – 11.15 Coffee / Tea
- 11.15 – 12.15 **T. Strick** (Cold Spring Harbor Labs):
Single-molecule analysis of DNA uncoiling
by a type II topoisomerase
- 12.30 – 17.00 Lunch and informal discussion
17.00 – 17.15 Coffee / Tea
- 17.15 – 18.15 **Ch. Schönenberger** (Nanocenter Basel):
Electrical and mechanical properties of carbon nanotubes
- 18.15 – 20.00 Posters

Wednesday, February 21st

- 8.45 – 9.45 **N.N.**
- 9.45 – 10.45 **V. Colvin** (Rice University):
Photonic Band Gap Materials
- 10.45 – 11.15 Coffee / Tea
- 11.15 – 12.15 **C. Bechinger** (Physics Department, Universität Konstanz):
Colloidal suspensions as model systems for cooperative phenomena:
Order through disorder
- 12.30 – 17.00 Lunch and informal discussion
17.00 – 17.15 Coffee / Tea
- 17.15 – 18.15 **M. L. Roukes** (Condensed Matter Physics, Caltech, Pasadena):
Nanomechanical Systems and Force Detection
- 18.15 – 19.15 **E. Frey** (Lyman Laboratory of Physics, Harvard University):
Models of Molecular Motors

Thursday, February 22nd

- 8.45 – 9.45 **T. Basché** (Institut für Physikalische Chemie, Johannes Gutenberg-Universität Mainz):
Microscopy and Spectroscopy of Single Nanoparticles II
- 9.45 – 10.45 **J. Y. Walz** (Department of Chemical Engineering, Yale University):
Measuring Colloidal Forces at the sub-picoNewton Scale using Total Internal Reflection Microscopy
- 10.45 – 11.15 Coffee / Tea
- 11.15 – 12.15 **M. Rief** (Universität München):
Myosin Motors
- 12.30 – 17.00 Lunch and informal discussion
17.00 – 17.15 Coffee / Tea
- 17.15 – 18.15 **J. Hafner** (Harvard University):
Nanomechanics of Carbon Nanotube Probes

Friday, February 23rd

- 8.45 – 9.45 **U. Steiner** (University of Groningen):
Harnessing film instabilities for 100 nm lithography
- 9.45 – 10.45 **N.N.**
- 10.45 – 11.15 Coffee / Tea
- 11.15 – 12.15 **N. N.**
- 12.30 – 17.00 Lunch and informal discussion
17.00 – 17.15 Coffee / Tea
- 17.15 – 18.15 **J. F. Stoddart** (Department of Chemistry and Biochemistry, University of California):
Devices Based on Interlocked Molecules
- 18.15 – 19.15 **A. N. Cleland** (Department of Physics, UC Santa Barbara):
Integrated Engineered Nanodevices and Nanoelectronics

Poster Contributions

The abstracts of all posters can be found in alphabetical order.

- C. Bräuchle, G. Seisenberger, T. Endress, M. Ried, M. Hallek:
Single Virus Tracing: Infectious Entry Pathway of a Single Dye-labeled Virus into a Living Cell
- M. Brunner, P. Leiderer, C. Bechinger:
Phase behavior of two-dimensional colloidal systems in the presence of periodic light fields
- F.W. Deeg, C. Seebacher, C. Hellriegel, C. Bräuchle, S. Altmaier, R. Jäger, P. Behrens:
Dynamics of Single Chromophores in Nanoporous Molecular Sieves
- Heiko Elsinger, Johannes Boneberg, Clemens Bechinger, Paul Leiderer:
Self-assembled polyelectrolyte multilayers as enabling technology
- A. Erbe, C. Weiss, W. Zwerger, and R.H. Blick:
A nanomechanical resonator shuttling single electrons at radio frequencies
- D. Fischer, P. Nielaba:
Phasendiagramm einer Modellegierung mit elastischen Wechselwirkungen
- M. George, B. Stein, C. Kirchner, M. Seitz, H. E. Gaub:
The Field-effect addressable Potentiometric Sensor – protective coatings and potential resolution
- S. Griessl, F. Jamitzki, R. Schloderer, M. Edelwirth:
Structure studies on self assembled trimesic acid monolayers
- Nolan Holland, Tilman Schäffer, and Hermann Gaub:
The Measurement of the Force Transduction Through Partially Extended Polymer chains by Molecular Force Spectroscopy
- Thorsten Hugel, Matthias Grosholz, Hauke Clausen-Schaumann, Andreas Pfau, Hermann Gaub, Markus Seitz:
Elasticity of single polyelectrolyte chains and their desorption from solid supports studied by AFM based single molecule force spectroscopy
- Christa S. Krämer and Thomas J. J. Müller:
Phenothiazines as Building Blocks for Novel Redox Active Molecular Wires
- R. Krautbauer, T.E. Schrader, H. Clausen-Schaumann, M. Rief, L.H. Pope, S. Allen, H. E. Gaub:
Single Molecule Force Spectroscopy of DNA Complexed with Drugs

- S. Kremmer, C. Teichert, E. Pischler, H. Gold, F. Kuchar and M. Schatzmayr:
Nanometer scale characterization of silicon dioxide thin films with conducting atomic force microscopy (C-AFM) in ultra high vacuum
- K. Mangold, R. Bubeck, P. Leiderer and C. Bechinger:
Magnetic Induced Phase Transition in Two-Dimensional Colloidal Systems
- Rainer Matzke, Ken Jacobson and Manfred Radmacher:
Mechanical Properties of Cells – AFM Studies
- B. Stein, M. George, H. E. Gaub:
Noise analysis on the Light-addressable-Potentiometric Sensor (LAPS)
- Wolfram Strepp:
Monte-Carlo-simulation of hard disks in a external periodic potential
- F. Trixler, M. Reiter and W. Heckl:
The Influence of specific bounding properties in organic adsorbates to imaging and nanomanipulation with STM
- A. Wehle, M. Benoit, R. Huss, H.E. Gaub:
Changes in Adhesion Properties of Cells Studied on Hematopoietic Stem Cells
- A. Zumbusch, C. Bräuchle, T. Hellerer, H. Scheer, P. Zehetmayer:
Studies of the Energy Transfer in Single Phycoerythrocyanin Molecules

Invited Lectures

Abstracts

Nanochemistry and Colloids: The Basics

Vicki Colvin
Rice University

The scientific and technological value of forming solids with nanometer dimensions will be the subject of most talks in this winter school. This lecture will focus on the first challenge faced by nanoscience, that of fabricating a nanostructure. This problem now occupies the attention of growing field known as 'nanochemistry'. The aim of the effort is to prepare nanostructures that can ultimately be handled in the solution phase. Because they can be recovered in so many different forms (powders, solutions, thin films) they can be studied by a range of different techniques. In addition, their relatively inexpensive processing produces large quantities of material which makes them most attractive for future commercial technologies. Though the interest in nanochemistry is intense, developing a robust chemical synthesis for a nanostructure remains one of the most challenging steps in nanoscience. In this talk we will cover the last 15 years of work on the chemical synthesis of nanocrystals, including core-shell semiconductor nanocrystals, metal and ceramic nanocrystals. The chemical strategies used to limit crystal growth in solution will be introduced, and the relative advantages of each method highlighted. Additionally, I will introduce the techniques that when taken together constitute a gold standard for the characterization of a nanocrystalline system. We'll finish the review with a summary of the current outstanding problems in the field.

Possibly relevant/interesting papers:

Alivisatos, A. P. (1996). "Semiconductor Clusters, Nanocrystals and Quantum Dots." Science **271**(February 16): 933-937.

Mirkin, C. A., R. L. Letsinger, et al. (1996). "A DNA-Based Method for Rationally Assembling Nanoparticles into Macroscopic Materials." Nature **382**: 607-611.

Murray, C. B., D. J. Norris, et al. (1993). "Synthesis and Characterization of Nearly Monodisperse CdE (E, S, Se, Te) Semiconductor Nanocrystallites." J. Am. Chem. Soc. **115**: 8706-8715.

Trentler TJ. Denler TE. Bertone JF. Agrawal A. Colvin VL. "Synthesis of TiO₂ nanocrystals by nonhydrolytic solution-based reactions." *Journal of the American Chemical Society.* 121(7):1613-1614, 1999 Feb 24.

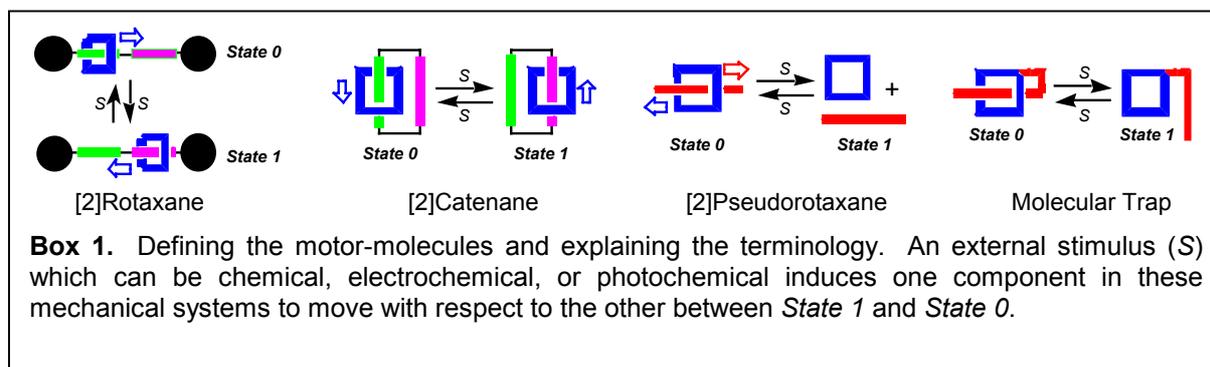
V. L. Colvin, M. C. Schlamp and A. P. Alivisatos, "Light Emitting Diodes Made with a Semiconducting Polymer and Cadmium Selenide Nanocrystals" *Nature* **370**, 354 (1994).

ARTIFICIAL MOLECULAR MACHINES

J Fraser Stoddart

*Department of Chemistry and Biochemistry, University of California, Los Angeles,
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Molecules with interlocked components, called rotaxanes and catenanes (**Box 1**) can now be synthesized¹⁻⁵ quite efficiently using template-directed protocols. Since the weak inter-component forces that guide their syntheses 'live on' inside the molecules afterwards, they can be activated such that their components move with respect to each other in either a linear fashion (e.g., the ring component along the rod of the dumbbell component of a [2]rotaxane) or a rotary manner (e.g., one ring of a [2]catenane circumrotating through the other ring). Thus, [2]rotaxanes can be likened to linear motors and [2]catenanes to rotary motors. Moreover, these molecules can be activated^{6,7} by switching the recognition elements on and off between the components with chemicals, electricity, or light, such that they will perform motions (e.g., shuttling actions or elongations or contractions) reminiscent of the moving parts in macroscopic machines. Such molecular motors and machines hold promise as the 'intelligent' building blocks for the fabrication of devices at the nanoscale level.



This lecture will describe recent progress⁸ toward the construction of photochemically-, electrochemically, and chemically-driven molecular machines based on supramolecular (pseudorotaxanes) and molecular (rotaxanes and catenanes) systems, including the molecular trap (**Box 1**).

1. "Interlocked and Intertwined Structures and Superstructures", *Chem. Rev.* **1995**, *95*, 2725-2828.
2. "Self-Assembly in Natural and Unnatural Systems", *Angew. Chem. Int. Ed. Engl.* **1996**, *35*, 1154-1196.
3. "Interlocked Macromolecules", *Chem. Rev.* **1999**, *99*, 1643-1663.
4. "Cyclodextrin-Based Catenanes and Rotaxanes", *Chem. Rev.* **1998**, *98*, 1959-1976.
5. "Molecular Catenanes, Rotaxanes and Knots", Sauvage, J.-P.; Dietrich-Buchecker, C., Eds., Wiley-VCH, Weinheim, **1999**.
6. "Molecular Machines", *Acc. Chem. Res.* **1998**, *31*, 405-414.
7. "Artificial Molecular Machines", *Angew. Chem. Int. Ed.* **2000**, *39*, 3349-3391.
8. For an example, see "A Photochemically-Driven Molecular-Level Abacus", *Chem. Eur. J.* **2000**, *6*, 3558-3574.

Single molecule spectroscopy : probing of charge transport at a nanometre scales

J.-M. Caruge and M. Orrit*

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The narrow optical transitions of organic molecules held in rigid matrices at low temperatures enable the detection and the spectroscopy of single molecules. Single molecules lines can be used to study the photophysics of the molecules, or their interaction with laser fields, as will be illustrated in the first part of the talk with a few examples [1]. Single molecules can also serve as ultrasensitive probes for the environment at a nanometre scale. Earlier work has focused on two-level system dynamics in amorphous matrices, and more recently on exciton migration dynamics in a molecular crystal. In the second part of the talk, we present recent experiments probing electric currents in a transparent, conducting semiconductor with single molecules deposited on the surface.

We deposited single terrylene and tetra-tert-butyl-terrylene molecules in thin hexadecane films on top of thin films (20 nm thickness) of indium-tin oxide doped with germanium oxide. After applying a static voltage to the semiconductor, we observed very strong quadratic shifts of the electronic transitions of single molecules. The magnitude of the shift was about 1 GHz at most, for an applied voltage of 1 V, corresponding to a current of about 1 mA in the film. The lines shifted more often to the red, but could also shift to the blue. Most molecules showed a well-defined threshold, above which a different regime occurred, with a slower increase, or sometimes even a decrease of the shift with voltage.

In order to probe the mechanism of the shift, we applied an ac-voltage with a variable frequency. To our surprise, we often observed well-defined resonances of the shift, in a frequency range between 10 kHz and 1 MHz. The magnitude, frequency, and width of the resonance all depended on the particular molecule investigated, even within the same illuminated spot of a few square microns. Some molecules showed self-oscillations of their transitions frequency for a sufficiently high applied dc-voltage, with oscillation frequencies as low as 100 Hz. In some cases, the self-oscillation frequency coincided with that of the resonance observed in the ac-voltage response. So far, we could not find any plausible mechanism for such slow processes associated with small length scales. We feel that electronic, acoustic or electromagnetic resonances would occur at much higher frequencies, and the elucidation of the origin of the resonances and self-oscillations requires further experimental and theoretical work.

From complementary experiments, we deduced that the observed shift is not an ordinary Stark effect, and that it is not caused by thermal effects in the lattice. However, we determined that, for the currents applied, the electronic temperature is significantly higher than that of the lattice. A change in the permittivity of the electron gas due to heating could explain the shifts of single molecule lines. If our interpretation is correct, the single molecule acts as an electric wattmeter on a nanometre scale. Such a component based on a single quantum system (be it an organic molecule, a semiconductor nanocrystal, or a self-assembled quantum dot) could be used in scientific applications or in future devices, to relay local information from the nano-world.

[1] see, for example: Ph. Tamarat et al., J. Phys. Chem. A 104 (2000), 1-16.

Nanomechanical Systems

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The ability to create mechanical nanostructures allows one to measure extremely small forces and masses. Nanofabrication approaches such as electron beam lithography and thin film processing techniques can be used to scale traditional micro-electromechanical systems from micrometer to nanometer dimensions. Such systems have been used as ultra-sensitive force transducers, measuring forces associated with single molecules, for example. Ultra-low mass high quality factor resonant systems can be used as highly sensitive mass sensors. We have explored new motion transduction methods such as modulated electron beams for interrogating the motion of mechanical systems with high spatial resolution. In many such systems the mechanical properties, surface effects and process-induced defects can strongly influence the properties of the devices. These materials issues must be considered when utilizing nanomechanical structures.

A different nanomechanical approach to probing and sorting individual molecules involves the controlled driving and motion of molecules through nanofabricated fluid systems. A variety of lithographic and self-assembled techniques can be used to create controlled fluid systems. With dimensions comparable to relevant molecular length scales, a variety of physical phenomena can be probed and used to sort molecules or nanoparticles. Relevant length scales include the molecular size or diffusion lengths in a fluid system. We will describe several such systems.

Nanofabrication approaches can also be used to produce surface chemical modifications at the nanoscale and to create mechanical devices that can be interfaced to functional molecules. Artificial mechanical nanostructures can be coupled to motor molecules, for example, to be used in transduction of molecular motion. Nanoscale modification of molecular monolayers can be used for the selective placement of structures by selective chemical binding. The simultaneous use of lithographically created nanostructures with biochemical activity presents new opportunities for studies and devices at the nanoscale.

Spinodal Decomposition of Colloids

J.K.G Dhont

Forschungszentrum Jülich

Colloidal systems are solutions of very large molecules (in the size range of a few nm to a few microns) in solution. These large molecules, the “colloidal particles”, exhibit appreciable thermal motion, and can therefore serve as model systems for molecular/atomic systems. There are, however, many features that are special for colloidal systems. Due to the very slow dynamics of the colloidal particles, colloidal systems show complex flow behaviour (like shear-thinning and thickening). Their large size allows for single particle manipulation and visualization. Moreover, there is an appreciable freedom in choosing a pair-interaction potential, which for molecular/atomic systems is more or less fixed. The extreme short-ranged and long-ranged interaction potentials give rise to phenomena that are not found in molecular/atomic systems, like gel-formation and flocculation.

This lecture will be concerned with a theoretical foundation of spinodal decomposition kinetics of colloidal systems. When a system is quenched into the unstable part of the phase diagram, density inhomogeneities develop, ultimately leading to coexistence of two bulk phases with different concentrations. The aim is thus to describe the temporal evolution of the spatially inhomogeneous density of colloidal particles. First of all, the Smoluchowski equation is introduced, which is an equation of motion for the probability density function of the slow phase space coordinates in the system of large colloidal particles and the solvent molecules. This equation of motion is the colloidal analogue of the Liouville equation for molecular/atomic systems. The Smoluchowski equation is then used to describe demixing kinetics in the initial stages. This is a microscopic foundation of the classic Cahn-Hilliard theory. The intermediate stage of demixing, which is described by a non-linear equations of motion for the density is also considered. In particular, dynamic scaling functions will be derived from the Smoluchowski equation, and compared to experimental results.

J.W. Cahn, J.E. Hilliard, *J. Chem. Phys.* **28** (1958) 258, **31** (1959) 688.

J.K.G. Dhont, *J. Chem. Phys.* **105** (1996) 5112.

J.K.G. Dhont, *An Introduction to Dynamics of Colloids*, Elsevier Science (1996), chapter 9.

Nanomechanics, adhesion and friction

B.N.J. Persson

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Nearly all macroscopic bodies have surface roughness on many different length scales. When two macroscopic bodies are in contact (e.g., a solid block on a substrate) the area of real contact consists of a large number of micrometer-sized contact areas, where the local pressure is so large that plastic yield usually has occurred. For macroscopic bodies the adhesive interaction between the bodies is usually unimportant, except for very smooth surfaces of elastically soft solids, e.g., rubber block on a smooth glass substrate.

For microscale (or nanoscale) sized objects the situation is completely different. First, only a few nanometer sized contact areas may occur, and the main interaction force between the block and the substrate is no longer the applied normal force (e.g., the load Mg), but the adhesion force will dominate. The latter force may have several contributions, e.g., the van der Waals force (which operates also between the non-contact surface area), or chemical forces, or the force from capillary bridges or from net charges generated, e.g., during sliding of the block relative to the substrate. Furthermore, on the nanometer scale solids are much harder than on the macroscopic scale, e.g., the plastic yield stress is typically a factor of 100 larger than on the macroscopic scale, and the solids in the contact areas may be elastically deformed rather than plastically deformed. In many modern applications, e.g., micromotors or disk drives, the adhesion force and hence the static friction force may be so large that the applied driving force may be too small to start the sliding motion. Furthermore, during sliding the strong adhesion force often results in a very large kinetic friction force, and accompanied large wear, and the sliding device may quickly wear out.

Nearly all surfaces of practical importance are lubricated. In fact, even if no lubrication fluid has been intentionally added to the system, a layer of organic molecules (greases) occur on most solid surfaces, since such layers form spontaneously on most hard solids by condensation from the atmosphere. I discuss boundary lubrication, and emphasize the fundamental role of the atomic corrugation of the confining solid walls on the squeezing and sliding of molecular thin lubrication films. The properties of atomic and molecular lubricants confined between two approaching surfaces are investigated by molecular dynamics and kinetic Monte Carlo calculations. In the limit of thin interfaces, the lubricant atoms form well defined layers, whose number decreases in discontinuous steps with increasing applied pressure. These transitions occur easily and completely for unpinned lubricant films, while they are sluggish and incomplete in the case of strong pinning. Before the transition, an intermediate phase arises, which facilitates the thinning of the lubricant. Lateral sliding of the surfaces enhances the thinning rate. I also show that for two-dimensional (2D) liquid-like layers, the squeeze-out exhibits instabilities which may result in trapped islands of lubrication molecules, as observed in recent experiments. Finally, I consider the sliding of boundary lubricated surfaces and discuss the nature of the lubrication film during the transition from stick to slip.

References:

Sliding Friction: Physical Principles and Applications,
Second (expanded) Edition (Springer, Heidelberg, June 2000).

Surface Science Reports 33, 1999, pp 83-120.

Microscopy and Spectroscopy of Single Nanoparticles I

Thomas Basché

Institut für Physikalische Chemie, Johannes Gutenberg-Universität Mainz

Experimental techniques for optical single molecule (nanoparticle) detection

Confocal fluorescence microscopy [1]

High resolution laser spectroscopy [1,2]

Fundamentals of semiconductor nanocrystals [3] and carbon nanotubes

- Preparation and structural properties
- Electronic and optical properties

References

[1] *Frontiers in Chemistry : Single Molecules*, Science (Special Issue), 283 (1999) 1667-1695.

[2] *Single Molecule Optical Detection, Imaging and Spectroscopy*, Eds.: Th. Basché, W. E. Moerner, M. Orrit, U. P. Wild, (VCH Weinheim 1997)

[3] A. P. Alivisatos, *Perspectives on the Physical Chemistry of Semiconductor Nanocrystals*, J. Phys. Chem. 100 (1996) 13226-13239

[4] S. A. Empedocles, R. Neuhauser, K. Shimizu, M. G. Bawendi, *Photoluminescence from Single Semiconductor Nanostructures*, Adv. Mater. 11 (1999) 1243.

Single-molecule analysis of DNA uncoiling by a type II topoisomerase

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Type II DNA topoisomerases (topo) are essential ATP-dependent enzymes capable of transporting a DNA through a transient double-strand break in a second DNA segment. This enables them to untangle replicated chromosomes and relax the interwound supercoils (plectonemes) which arise in twisted DNA. Here we present a micromanipulation experiment in which we follow in real-time the action of a single *D. melanogaster* topo II acting on a linear DNA which is mechanically stretched and supercoiled. By monitoring the DNA's extension, we directly observe the relaxation in the presence of ATP of two supercoils during a single catalytic turnover. By controlling the force pulling on the molecule we find that enzyme turnover decreases as the force acting on the DNA increases. This suggests that resealing the cleaved DNA is a rate-limiting step in the enzymatic cycle. Finally, in the absence of ATP, we observe the clamping of a DNA crossover by a single topo II on at least two different time-scales, i.e. configurations. These results demonstrate that single molecule experiments are a powerful new tool for the study of topoisomerases and other proteins which interact with DNA.

Electrical and mechanical properties of carbon nanotubes

Christian Schönenberger

Nanocenter Basel, Institute of Physics, Klingelbergstrasse 82, Ch-4056 Basel, Switzerland

The first part of my lecture will be a tutorial introduction into the physics of carbon nanotubes. I will emphasize the electronic properties for an ideal carbon nanotubes deduced from the simplest possible theory, describe the way nanotubes are produced and then summarize a few key experiments. In the second part of my talk, I will focus on the electrical properties of multiwalled carbon nanotubes and discuss recent experiments. In particular, I will demonstrate that nanotubes are certainly not perfect and as simple as tight-binding calculation suggests. On the one hand, nanotubes were recently found to be quite sensitive to an oxidizing environment (air, water) causing doping. The doping level can be substantial, in particular for larger diameter tubes. Tunneling experiments, on the other hands, have revealed pronounced zero-bias anomalies, both in single and multiwall carbon nanotubes. It is believed that this anomaly is a consequence of non-Fermi liquid behavior. I will also highlight a few experiments on the mechanical properties of nanotubes, in particular on those measuring the Young's modulus. Finally, if time permits I would like to discuss the status of electrical measurements on single DNA molecules.

I acknowledge contributions from the group of Laszlo Forro (EPFL), A. Bachtold, M. Buitelaar, M. Calame, F. Dewarrat, H.-W. Fink, K. Furukawa, M. Iqbal, T. Nussbaumer and C. Strunk.

Molecular Electronic Devices

Brosi Hasslacher

Departments of Electrical Engineering and Applied Physics, Yale University

Electron transport studies in molecular-scale systems have recently become possible with the utilization of advanced microfabrication and self-assembly techniques. We have performed the measurement of the conductance of a single molecule using a break junction technique [1], the demonstration of molecular diodes [2], and the systematic investigation of metal-molecule contacts in a variety of systems [3]. Most recently, we have observed [4] large and useful reversible switching behavior in an electronic device that utilizes molecules as the active component, specifically a nitroamine redox center. The molecular device exhibits negative differential resistance (NDR) and peak-to-valley ratios exceeding 1000:1 at low temperature [4] (which exceeds that observed in typical solid state quantum well resonant tunneling heterostructures) and room temperature operation [5]. The designs of molecular switches, memories [6], and their circuit applications will be discussed.

1. M. A. Reed *et al.*, *Science* **278**, 252 (1997).
2. C. Zhou *et al.*, *Appl. Phys. Lett.* **71**, 611 (1997).
3. J. Chen *et al.*, *Chem Phys Lett* **313**, 741 (1999).
4. J. Chen *et al.*, *Science* **286**, 1550 (1999).
5. J. Chen *et al.*, *Appl. Phys. Lett.*, **77**, 1224 (2000)
6. M. A. Reed, *Appl. Phys. Lett.*, in press.

Photonic Band Gap Materials

Vicki Colvin
Rice University

Photonic band gap materials are, quite simply, strongly diffractive solids with a characteristic length scale on the order of hundreds of nanometers. Because of their strongly diffractive behavior, they can in theory provide exquisite control over the photonic density of states in a solid. This talk will focus on the basic theory behind these systems, with an emphasis on developing a physical intuition for material design principles. The systems of interest will be based on the natural motif of the gemstone opal. This form can be replicated in the laboratory, providing opaline films which can be used to build up complex macroporous architectures. These porous materials consist of a regular array of holes arranged in a close-packed array. The optical properties, and the important role that defects and sample format play, in these systems will be outlined. Finally, we'll discuss the many possible applications for these systems, especially those that involve the combination of photonic band gaps with nanoscale materials.

(1) "Monodisperse Colloids and Their Colloidal Crystals" P. Jiang, J. F. Bertone and V. L. Colvin, to appear in *Science*, January 2001.

(2) "Template-directed preparation of macroporous polymers with oriented and crystalline arrays of voids" Jiang P. Hwang KS. Mittleman DM. Bertone JF. Colvin VL. *Journal of the American Chemical Society*. 121(50):11630-11637, 1999 Dec 22.

(3) "Porous metals from colloidal templates" Kulinowski KM. Jiang P. Vaswani H. Colvin VL. *Advanced Materials* 12(11):833-838, 2000 Jun 2.

(4) "Thickness dependence of the optical properties of ordered silica-air and air-polymer photonic crystals" Bertone JF. Jiang P. Hwang KS. Mittleman DM. Colvin VL. *Physical Review Letters*. 83(2):300-303, 1999 Jul 12.

Colloidal suspensions as model systems for cooperative phenomena: Order through disorder

Clemens Bechinger

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Colloidal suspensions consist of mesoscopic particles which are suspended in fluid mediums like water or alcohols. Similar to atomic solids, crystallization, melting, and glass transitions can also be observed in colloidal suspensions. However, since the typical length scales in colloidal suspensions are in the micrometer range, optical methods like static and dynamic light scattering, or light microscopy can be employed to study structure and dynamics of such systems. Colloidal suspensions allow detailed insight in physical processes (e.g. formation and growth of crystals or glasses) which are not always accessible with such a precision on the atomic scale. Consequently, colloids are regarded as model systems and can substantially contribute to the understanding of processes in atomic systems but also elucidate problems in the context of statistical physics.

Using colloidal suspensions we first discuss the origin of forces which are entirely caused by entropy. Such entropic forces are crucial for the understanding of phase separation phenomena in polydisperse particle mixtures but are also discussed in the context of biological systems. Secondly, we study the phase behavior of a two-dimensional colloidal system in the presence of a periodic light field. Under the influence of light forces the dielectric spheres are driven into regions of highest light intensity I . When I is increased the particles' fluctuations perpendicular to the lines are decreased and one observes a transition from a disordered into an ordered state which is known as light-induced freezing. However, upon further increase of the intensity, i.e. upon further reduction of fluctuations perpendicular to the light potential, the crystal is found to remelt (laser-induced-melting) to a 1D liquid. We present a simple model which explains this intriguing phenomenon in terms of particle fluctuations which tend to stabilize the crystalline phase. In addition we also point out the relevance of the observed effect for other systems.

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Nanomechanical Systems and Force Detection

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Nanomechanical systems offer the prospect of unprecedented sensitivity, bandwidth and spatial resolution for applications ranging from sensors to explorations of fundamental physics. In this presentation I will review the principles of force detection by nanomechanical systems, both in vacuum and in fluid. I will discuss some of the most exciting possibilities for their use – among which are magnetic resonance force microscopy and single-molecule force assays – and review the state of the art in these fields (including the ongoing work in my group). Finally, the prospects for attaining quantum-limited measurements with nanomechanical systems will be explored.

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Models of Molecular Motors

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Motor proteins are enzymes with the ability to transform chemical energy, derived from the hydrolysis of adenosinediphosphate (ATP), into mechanical work. This allows these nano-scale machines to move along molecular tracks and perform various transport tasks relevant for a wide range of biological processes, e.g. cell division, muscle contraction, and intracellular transport of organelles. For many decades exclusively data from physiological measurements on muscles provided experimental information for modeling molecular motors. In recent years, a variety of *in vitro* techniques allowed the observation of single motor proteins with an extremely high spatial and temporal resolution and gave new insights into the basic principles underlying their operation. This new generation of experiments sparked off an intensive interest in a theoretical description of molecular motors.

From a theoretical point of view molecular motors are isothermal stochastic processes with coupled chemical and mechanical cycles. The key features in these theoretical models are the following. (i) There are transitions between several well defined states of the enzyme. In each state the particle performs Brownian motion in a given potential landscape. The dynamics of the transition between the states is stochastic and typically described in terms of master equations. (ii) Unidirectional motion can be obtained in several ways, e.g., by asymmetries in the transition rates, specific conformational changes in the motor enzyme or various other mechanisms, which break the detailed balance of the stochastic processes.

In this lecture I will explain some elementary aspects of the theoretical description of molecular motors focusing on simple two-state models. The general ideas underlying theoretical concepts such as master equations, stochastic processes and numerical simulation will be introduced and discussed.

Some reviews for further study:

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Some useful links on the web:

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- [3] <http://www.cnr.berkeley.edu/~goster/home.html>

Microscopy and Spectroscopy of Single Nanoparticles II

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Applications of optical single molecule microscopy and spectroscopy [1,2]

- Single molecule optical switching
- Fluorescence microscopy of single semiconductor nanocrystals [4]
- Attempts to correlate structural and spectroscopic properties of single nanoparticles

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Measuring Colloidal Forces at the sub-picoNewton Scale using Total Internal Reflection Microscopy

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Total Internal Reflection Microscopy (TIRM) is a relatively new optical technique that can be used to study the motion of a single colloidal particle next to a solid plate in liquid solutions. In the technique, a continuous wave laser beam is made incident on the solid/liquid interface at an angle above the critical angle, such that an evanescent wave is produced in the liquid. The evanescent wave is unique in that the wave vector is real in the direction parallel to the interface yet imaginary normal to the interface. Thus the magnitude of the electric field of the wave decays exponentially away from the interface with a characteristic decay constant that is determined by the wavelength of incident light, refractive indices of the plate and liquid, and incident angle. For TIRM experiments, this decay length is typically around 100 nm. If a colloidal particle is located sufficiently close to the interface, the wave is scattered with an intensity that varies exponentially with the separation distance between the particle and plate. Thus measuring the intensity provides a sensitive and near-instantaneous method for determining separation distance.

The largest use of the technique to-date has been to measure the interaction energy profile between a single particle and plate. For this measurement, a Brownian particle is typically levitated above the plate by electrostatic repulsive forces between the charged surfaces. The particle, which is also acted upon by gravity and possibly other attractive colloidal forces, is trapped spatially in a local energy well. The probability distribution of separation distances sampled by the particle is described by Boltzmann's equation, which relates this probability to the interaction potential energy of the particle. Thus monitoring the separation distances for sufficiently long periods of time allows determining the particle/plate interaction energy profile. The method is sensitive enough to resolve energies produced by sub-picoNewton forces.

The technique can also be used to determine parameters describing the dynamics of the particle motion, such as the hindered mobility or diffusion coefficient. More information about the uses of the device can be found in the following articles.

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Myosin Motors

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Myosins are the motors that drive muscle contraction. However, over the last years a 15 different classes of myosins have been identified that are responsible for a variety of transport processes ranging from vesicle transport to cell division. The energy source for myosin movement is Adenosine Triphosphate (ATP). How the chemical energy of ATP-hydrolysis is converted into mechanical movement is still an open question.

In the last years single molecule studies using optical traps and glass microneedles have allowed a mechanical characterization of single myosin molecules. The step size of the motors could be measured directly. Such measurements have shown that different members of the myosin family have different chemo-mechanical properties depending on their respective physiological roles. Muscle myosin works in large assemblies that are able to produce rapid contractions. Each individual muscle myosin is attached to its actin track for only a short part of the ATP cycle but cannot move continuously over larger distances. Therefore, muscle myosin is a non-processive motor. The vesicle transporter myosin V, on the other hand, has to move vesicles over larger distances in a cell. Using an optical trap myosin V could be identified as a processive motor that moves continuously in 36 nm steps along the actin helix. A statistical analysis of the time distributions of myosin V steps reveals important details of the ATP hydrolysis cycle: The release of ADP from the myosin-V molecule is the rate limiting process of the cycle and myosin V uses one ATP molecule per step.

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Nanomechanics of Carbon Nanotube Probes

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Carbon nanotubes are fascinating one-dimensional structures. They consist of single or multiple graphene sheets rolled into tubes with only nanometer-scale diameters, yet lengths of many microns. The nanomechanics of carbon nanotubes have received considerable attention due to their

remarkable properties. Nanotubes are extremely stiff, with an observed Young's modulus of approximately 1.2 TPa [1], and have a tensile strength of about 50 GPa [2]. Their unique structure allows them to elastically buckle rather than fracture under large loads, making them highly robust to mechanical deformations [3]. Studies of nanotubes sliding and rolling on surfaces have elucidated these microscopic processes [4], and measurements of the inter-wall sliding friction in nanotubes suggest their use as nanometer-scale bearings [5]. The experimental techniques employed to make these measurements and their results will be reviewed. The unique structure and properties of carbon nanotubes make them ideal probes for atomic force microscopy (AFM) [6]. We have recently developed simple methods to fabricate single-walled nanotube AFM tips in high yield [7]. Several nanomechanical properties of carbon nanotubes, such as thermal vibrations, elastic buckling, and sliding friction, determine the optimum nanotube tip structure for robust, high-resolution probes. The fabrication, nanomechanical considerations, and performance of these nanotube AFM tips will be presented.

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Harnessing film instabilities for 100 nm lithography

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Cutting-edge semiconductor technology, it is based on a conceptually simple principle - the optical projection of a circuit pattern on a photosensitive polymer film, and the subsequent etching of the pattern into the silicon chip. With current structure sizes of 100-200 nm, a further reduction of lateral length scales using optical process is becoming increasingly difficult [1]. Alternative processes based on non-optical techniques may be an attractive alternative for higher integration densities on semiconductor devices.

In my talk I will focus on two aspects: (1) the fundamentals of hydrodynamic instabilities in thin film, and (2) controlled pattern formation employing these instabilities.

In the absence of external or dispersion forces, thin liquid films are intrinsically stable. In practice, however, films are found to break-up into drops. I will discuss the physics of the hydrodynamic instabilities which cause film break-up [2] and will give two examples for external forces that cause such an instability: electrostatic [3,4], and thermomechanical forces [5]. As opposed to Van der Waals forces, which may also cause films to dewet, the application of an external potential allow a direct control of the instability. In particular, the availability of an external control parameter (e.g. voltage) allows the systematic study of thin film instabilities.

This control by external potential can be employed to transfer a master patter into a polymer film. Structure sizes from tens of micrometers down to 100 nm can be reliably replicated [3,5].

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Kinesin Motor Proteins

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Kinesins are motor proteins that move along microtubule filaments (1). Based on sequence features of the motor domain as well as other regions, the kinesin superfamily comprises at least 8 families. The common denominator of kinesin motors is the catalytic motor domain. Based on sequence comparisons, it possesses a P-loop type ATP binding site and a number of signature sequences only found in kinesins (2). Some of the latter are now known to be responsible for the interaction with the microtubule, others are of unassigned function.

The atomic structure of the kinesin motor domain reveals a remarkable structural homology with myosin and G-proteins, which also are P-loop nucleotidases. This suggests that the catalytic core domain plays a similar role in these three classes of proteins: it appears to be an engine that exploits the hydrolytic energy of ATP's phosphodiester bond to drive a conformational change. In kinesin motors this is controlled by the interaction with the microtubule. Free in solution, the ATPase is inactive and the molecule rests in its ADP form. The hydrolytic cycle can only be initiated upon binding to the microtubule, a process resembling the activation of myosin by actin, or the activation of G-proteins by their respective activators and exchange factors. However, based on this description, the catalytic domain is no more than an allosteric enzyme, and not a motor protein. Additional components are required. In conventional kinesins, the neck linker and neck domains, and probably also the hinge domain, cooperate with the motor domain to render the molecule fully motile. By analogy with muscle myosin one might speculate that these additional parts constitute a lever that amplifies the conformational changes in the core motor domain to give rise of the 8 nm steps observed in bead assays. This model, however, does not account for a feature of motility first observed in kinesins: processive motility of single motor molecules. Conventional kinesin is capable of stepping several hundreds of steps along the filament without dissociating, implying a tight coupling between two motor domains that are held together by virtue of the neck domain (3,4). While one of the motor heads moves to a forward microtubule binding site the other one remains bound to the 'track' and prevents dissociations of the molecule. The exact nature of this head-head interaction is the main objective of the current research.

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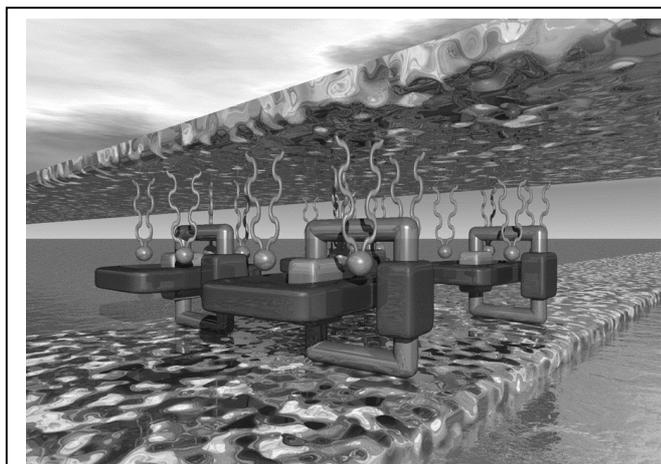
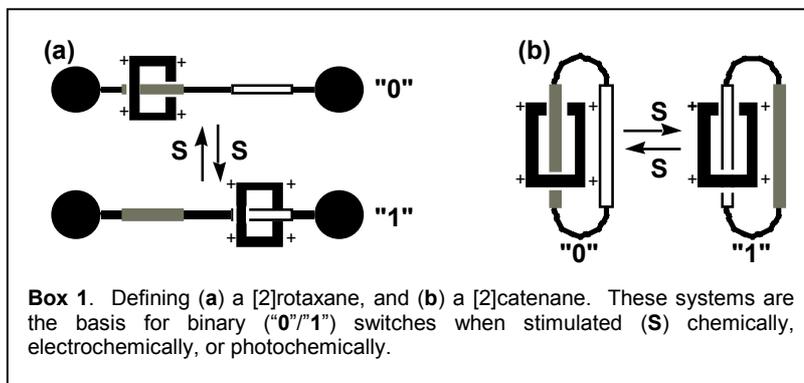
DEVICES BASED ON INTERLOCKED MOLECULES

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The area of molecular electronics has advanced considerably during the past few years. While there are several reasons for this progress, one enabling factor has been the rapid development in methods for synthesizing, not only bistable supramolecular complexes, but also bistable molecular compounds. Now the time is ripe to exploit these advances in both noncovalent and supramolecularly-assisted covalent synthesis¹ in the

context of solid-state device fabrication. Over the past couple of years, we have utilized a number of electrochemically switchable molecular and supramolecular systems as the active elements in solid-state switchable tunnel junction devices. In this lecture, the template-directed synthesis² and characterization of some bistable, mechanically-interlocked molecular compounds (**Box 1**) – catenanes and rotaxanes³ – will be discussed, prior to giving an account of the properties of devices that can be fabricated using this unique class of compounds.



Box 2. Recent work in our laboratories has focused on the synthesis of a particular [2]catenane composed of a tetracationic cyclophane interlocked by a macrocyclic polyether which incorporates a tetrathiafulvalene (TTF) unit and a 1,5-dioxynaphthalene (DNP) ring system. The ground state of the switch in solution involves the cyclophane encircling the TTF unit. Upon chemical or electrochemical oxidation of the TTF, the crown ether circumrotates such that the DNP station becomes encircled by the cyclophane. A solid-state, electronically addressable, bistable [2]catenane-based molecular switching device has been fabricated³ from a single monolayer of this [2]catenane by anchoring it with phospholipid counterions, prior to sandwiching it between an *n*-type polysilicon bottom electrode and a metallic top electrode. The device exhibits hysteretic (bistable) current/voltage characteristics. The switch, which is opened at +2 V, closed at -2 V, and read at +0.2 V, may be recycled many times under ambient conditions. A chemomechanical mechanism for the action of the switch in the solid-state will be presented and shown to be consistent with temperature-dependent measurements of the device operation.

An architectural rationale and an experimental program aimed at the development of molelectronic switching devices for memory and computing applications is discussed. Two-terminal molecular switch tunnel junctions are identified as the critical device components of molecular electronics-based circuitry that can both tile in two-dimensions, and are tolerant of manufacturing defects. Singly- and multiply-configurable solid state switching devices that are based upon electrochemically-switchable molecular (**Box 2**) and supramolecular systems are discussed in terms of both the synthesis of the molecular components,⁴ and the fabrication and performance of the devices.⁵

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Integrated Engineered Nanodevices and Nanoelectronics

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We have been pursuing the development of nanoscale mechanical structures that are integrated with superconductive electronic devices, including normal metal-insulator-superconductor (NIS) tunnel junction circuits and single-electron transistors. The goal of our research is to develop integrated, mechanically active structures with quantum-limited displacement and energy sensitivity. We are developing these types of devices for applications in single-photon limited far-infrared bolometry, on-chip electronic refrigeration, and quantum-limited motion sensing.

We are focussing on the use of GaAs single-crystal substrates for the mechanical structures. Heterostructures including an AlGaAs sacrificial underlayer are patterned and etched using standard nanomachining technology. The mechanical structures range from simple $0.1 \times 0.1 \times 10 \mu\text{m}^3$ beams with tunnel junctions defined on the top surface, to $1 \times 1 \times 0.2 \mu\text{m}^3$ blocks for bolometric and calorimetric applications, to beams with interdigitated capacitive coupling structure for capacitive displacement sensing.

Our bolometer designs include integrated SINIS tunnel junctions, whose low-voltage conductance depends exponentially on temperature below the superconducting transition temperature, yielding nanoscale electron thermometry. When biased just below the superconducting gap such circuits also allow evaporative cooling of the normal-metal electrons, which can then be cooled to below the temperature of the ambient phonons. The bolometers include two such circuits, allowing cooling and thermometry on a single, suspended nanoscale structure. The heat capacity of such a structure is of order $10^3 k_B$ at 100 mK, with thermal time constants of a few tens of microseconds, making these ideal for infrared bolometry.

Our mechanically active structures include integrated single-electron transistors as displacement sensors, which promise the possibility of near quantum-limited motion detection for radiofrequency resonators. The coupled resonator structures have fundamental resonances in the 10-100 MHz range, and can be driven separately from the detection circuitry.

I will discuss our implementations and present status for the progress in these two main thrusts of our research. The research has been pursued in collaboration with C.S. Yung, J.S. Aldridge, R.S. Hoy, Dr. R. Knobel and Dr. D. Schmidt. Support has been provided by ARO, NASA, NSF, IGPP and the Research Corporation, as well as funds from the University of California and QUEST.

Posters

Abstracts

SINGLE VIRUS TRACING: INFECTIOUS ENTRY PATHWAY OF A SINGLE DYE-LABELED VIRUS INTO A LIVING CELL

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Single molecule experiments were conducted to follow the migration of single adeno-associated viruses on their infectious entry pathway into living HeLa cells. Only one fluorescent dye molecule (Cy5) was attached to the viruses in order not to influence their physiological behavior. For the first time we could obtain diffusion trajectories of *single* viruses in four different stages of the infection:

- 1.) Diffusion of the virus in front of the cell membrane and receptor mediated adsorption
- 2.) Formation of the endosome with inclusion of the virus
- 3.) Transport of the endosome in the cell plasma and release of the virus
- 4.) Penetration and diffusion of the virus in the cell nucleus

From the trajectories of the individual viruses diffusion coefficients were obtained for all four stages. A detailed picture of the processes involved was modeled. Adeno-associated viruses show promising prospects for the use in human gene therapy. For this purpose a detailed understanding of the interactions of the virus and the target cell is important.

Phase behavior of two-dimensional colloidal systems in the presence of periodic light fields

M. Brunner, P. Leiderer, C. Bechinger

We investigated the phase behavior of a two-dimensional suspension of charge stabilized polystyrene spheres in the presence of a one-dimensional periodic light field. With increasing light intensity we observe a liquid-solid followed by a solid-liquid transition which is known as laser-induced freezing and melting, respectively. Here we report about measurements where in addition to the light intensity also the single particle density was systematically varied. As a result, we obtain for the first time the full thermodynamic information about the system which allows comparison with numerical predictions of other authors.

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DYNAMICS OF SINGLE CHROMOPHORES IN NANOPOROUS MOLECULAR SIEVES

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The last decade has seen a dramatic development in the synthesis of so-called molecular sieves, inorganic crystals with pore systems on the nanometer scale. At the same time it has been demonstrated that organic guests can be incorporated into these hosts in a well-defined manner, and supramolecular structures with unique degrees of freedom can be realized. In this contribution we will present the first spectroscopic study of individual chromophores in various porous host matrices by confocal fluorescence microscopy.

For example we have measured the orientation of individual oxazine-1 chromophores in the one-dimensional channel system of an AlPO₄-5 crystal. Most molecules are aligned along the channels and show no signs of rotational mobility. A minority of molecules, however, is found at odd orientations with the macroscopic channels and exhibit reorientational jumps on a second timescale. We assume that the latter kind of molecules is situated in local defects with extensions large than the size of the guest chromophore.

In another example we have introduced terylenediimide (TDI) molecules into the larger channels of a non-calcined tenside-filled monolithic MCM41 host. Whereas a few molecules exhibit a behaviour similar to the Oxazine-1/AlPO₄-5 sample, most molecules are mobile and show translational motion within the channel system. Whereas the diffusion in the pure tenside is characterized by a single diffusion coefficient, the chromophore mobility in the restricted environment of the porous host exhibits two slightly different coefficients and is about two orders of magnitude slower.

We will also discuss the potential of this novel class of materials for molecular switches/devices based on the dynamics of individual molecules in tailor-made nano-dimensioned vessels.

Self-assembled polyelectrolyte multilayers as enabling technology

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Ultrathin polymer films fabricated by electrostatic layer-by-layer¹ (LBL) assembly open scopes in designing constructed materials. Recent developments show a growing number of applications in optics, electronics and biotechnics. Here, we suggest the application of such multilayers as insulating spacer films in thin film transistors.

The alternating adsorption of polyanions and polycations on charged surfaces is a easy to use system and has been demonstrated to work in wide range of processing parameters.

We investigate the layer-by-layer adsorption and the adsorption kinetics by means of surface plasmon spectroscopy which allows to resolve a linear increase in layer thickness with the number of polymer layers. The layer thickness is mainly controlled by the amount of salt added to the polyelectrolyt aqueous solution. In additon we investigated the porosity and roughness of the films by means of atomic force microscopy. Our results show that only very thin films are free of defects whereas larger numbers of monolayers contain substantial amounts of defects.

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A nanomechanical resonator shuttling single electrons at radio frequencies

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We observe transport of electrons through a metallic island on the tip of a nanomechanical pendulum. In this experiment we combine mechanical motion with electronic properties on a nanometer scale. This combination can be a good candidate for a current standard, if experiments are done in a Coulomb-Blockade-regime. We show measurements on such a nanomechanical resonator shuttling single electrons at radio frequencies.

The resulting tunneling current shows distinct features corresponding to the discrete mechanical eigenfrequencies of the pendulum. We report on measurements covering the temperature range from 300 K down to 4.2 K. We explain the I-V curve, which differs from previous theoretical predictions, with model calculations based on a Master equation approach. The main difference to common single electron transistor devices is the fact that only one tunneling barrier is open at a certain time. This leads to an exponential suppression of cotunneling effects and thus increases the accuracy of current transport. Calculations for theoretical limits of the accuracy show that these devices will allow measuring quantum fluctuations.

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Phasendiagramm einer Modellegierung mit elastischen Wechselwirkungen

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Der Einfluss elastischer Wechselwirkungen auf das Phasendiagramm einer zweidimensionalen binären Modellegierung mit antiferromagnetischen Naechste-Nachbar- und ferromagnetischen Uebernaechste-Nachbar-Wechselwirkungen, wird anhand von Monte Carlo Simulationen und Techniken des Finite-Size-Scaling untersucht. Die elastischen Wechselwirkungen fuehren, im Vergleich zum Modell ohne elastische Effekte, zu einer Verringerung der kritischen und trikritischen Temperatur.

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The Field-effect addressable Potentiometric Sensor – protective coatings and potential resolution

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One of the first collaborations within the framework of CeNS between the labs of semiconductor- and biophysics dealt with idea to employ a novel surface potential sensor for cell-semiconductor hybrids. The main feature of the proposed field-effect addressable potentiometric sensor (FAPS) is the possibility to obtain high spatial resolution by a grid wise arrangement of field-effect channels and underlying gate-electrodes. By locally depleting the charge carriers in one field-effect channel via biasing one single gate electrode the channel resistance increases and is selectively sensitive to potential changes below AND above the cross-section of the addressed channel/gate electrode. In this manner surface potential changes induced by extracellular potentials of adhered cells can steer the channel resistance and therefore be detected as modulations of the voltage drop along the channel, if the FAPS is driven in a constant current mode. The technological challenge for the realization of the FAPS is the composition of conducting (metallic) gate electrodes and perpendicular (semi-conductive) field-effect channels lying on top. One promising method for the preparation of that kind of layered structures is the technique of the so-called epitactical lift-off (ELO) of GaAs-heterostructures that then can be transferred onto almost any desired pre-structured substrate. For the use of these ELO-films as component in a cell-sensor three essential problems have to be solved: i) Whereas for most applications the van-der-Waals interactions are sufficient to stabilize the GaAs-ELO-films on e.g. glass-substrates, the films tend to detach from the surface, when the devices are incubated in electrolytic solutions for several days. ii) Without a appropriate coating of the GaAs-films arsenic ions go into solution and poison the cell media. iii) In electrolytic solutions, like they are vital for cells (i.e. 140 mM NaCl), an enhanced etching of uncovered GaAs-surfaces can be observed, that affects the electrical properties of the field-effect channels. These facts prove the need for coatings that support the stability of the ELO-films, while being thin enough to allow for effective electrical coupling between the cells and the field-effect channels. In this work several basic approaches (evaporated inorganic insulators (SiO₂, Al₂O₃), low temperature PECVD-layers (Si₃N₄, SiO), spin-coated PMMA and thiol-based polysiloxan-layers) are investigated with respect to biocompatibility, electrical insulation and stabilization of the ELO-films. In a second part the fundamental limitations for the potential resolution of the FAPS are discussed. Especially the question, if the pooling of several addressable sites to one field-channel causes a loss of sensitivity compared to a single field-effect transistor and the dependence of the potential resolution on the size of the active regions is addressed.

Structure studies on self assembled trimesic acid monolayers

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The adsorption of trimesic acid (TMA) on HOPG and Ag(111) has been studied in ambient conditions and UHV. This work focuses on inducing this particular self assembly mechanism of hydrogen bonded organic molecules on crystal surfaces. STM, TDS and LEED measurements show the two dimensional self assembled adsorbate layers as predicted from the bulk model. Both STM and LEED measurements allow together a suggestion of the size and configuration of the adsorbat unit cell.

The Measurement of the Force Transduction Through Partially Extended Polymer Chains by Molecular Force Spectroscopy

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The dynamics of force transmission through single extended polymer chains were investigated using single molecule force spectroscopy. Partially extended polymer chains (DNA or polysaccharides) were fixed between an atomic force microscope probe and a substrate. The substrate was then driven to stretch the polymer and the propagation of the tension through the chain was observed by the AFM force sensor. Force sensors were small AFM cantilevers with low spring constants and relatively high resonant frequencies to maximize the sensitivity and time resolution. Two different means of driving the substrate were employed: input of a step wave and driving the surface with a sine wave. For both polymer systems, it was observed that when the step offset is applied, there is a force transduction which occurs faster than our time resolution. Beyond this, two distinct relaxations in the DNA were observed, with characteristic time constants of around 50 ms and 500 ms. No corresponding relaxations were observed in the polysaccharide system, which behaved in an elastic manner. Driving the DNA with a sine wave, it was observed that there was an observable phase shift in the force response from the sample displacement. The phase offset is dependent on chain extension and frequency of the oscillation. These results indicate that on the accessible time scales, the DNA molecules show dissipative (viscous) and conservative (elastic) behavior while the polysaccharides show only elastic response.

**Elasticity of single polyelectrolyte chains and their desorption from solid supports studied
by AFM based single molecule force spectroscopy**

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Abstract.

AFM based single molecule force spectroscopy was used for the investigation of single polyelectrolyte chains. Namely, the elasticity of polyvinylamine chains and their desorption from solid surfaces was studied as a function of the polymer's charge density and electrolyte concentration. Experimental force-distance profiles were fitted by the worm like chain model, including elastic contributions arising from the stretching of bond angles and covalent bonds. It was found that under the high stretching forces which can be applied in the AFM experiments, the bending rigidity of polyelectrolyte chains (as described by the persistence length) is significantly lower than predicted by Odijk-Skolnick-Fixman (OSF)-theory. Furthermore, the desorption force of single physisorbed polymer chains from negatively charged silica surfaces was determined. In addition to the electrostatic interaction between polymer and substrate, which depends linearly on the Debye screening length and the polymer's line charge density, a constant non-electrostatic contribution to the desorption force was observed.

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Phenothiazines as Building Blocks for Novel Redox Active Molecular Wires

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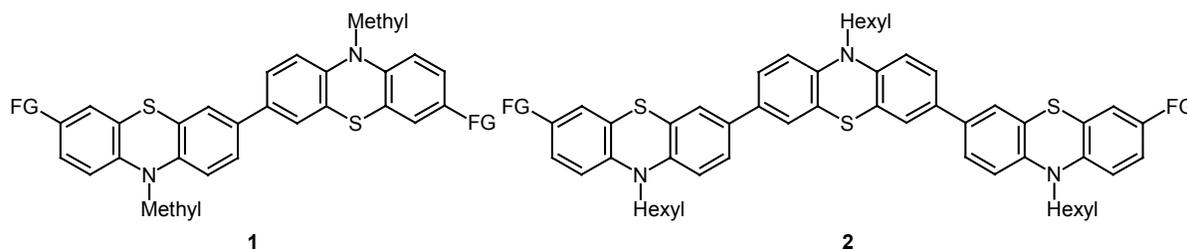
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Phenothiazines are electron rich, tricyclic heterocycles^[1] with interesting electro- and photochemical properties. Due to reversible oxidation phenothiazine derivatives have become attractive electrophor motifs in supramolecular and materials sciences.

Recently, we have found a straightforward access to alkynylated oligophenothiazines^[2] that has been extended to the synthesis of diphenothiazinyl dyads (**1**) and triphenothiazinyl triads (**2**). Synthetically, functional groups attached to the electro-active moiety are crucial as links to extended conjugated π -systems, i.e. molecular wires.



FG: functional group, i.e. CHO, Hal, Alkynyl

Cyclic voltammetry, absorption and emission spectroscopy studies reveal that the phenothiazine units in **1** or **2** are electronically coupled. First scanning tunnel microscope (STM) experiments now disclose an insight in self-organization potential of these molecules. The electronic properties and nanostructuring of systems like **1** and **2** will be presented and discussed.

Future goals will be the incorporation of these redox dumbbells into conjugated oligomers to furnish a new class of redox addressable molecular wires^[3] with lengths between 5-20 nm. Ultimately, the conductive state of these single molecules shall be generated by a photoinduced reversible electron transfer.

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Single Molecule Force Spectroscopy of DNA Complexed with Drugs

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Individual doublestranded DNA molecules undergo a highly cooperative structural transition when they are stretched with forces of 65 - 70 pN, leading to an overstretched structure (S-DNA). If the molecules are stretched with higher forces a second structural transition (melting transition) is observed, in which the double strand is mechanically separated, leaving only a single strand attached between AFM-tip and substrate. These well known features were used to characterize the changes in DNA mechanics upon binding of different drugs.

Cisplatin can introduce intrastrand and interstrand crosslinks, preferentially between guanine bases. Stretching experiments showed that the cooperativity of the BS-transition is reduced in all DNA molecules with bifunctional cisplatin adducts. In poly(dG)opoly(dC) and poly(dG-dC)opoly(dG-dC) DNA the drug inhibits the high force melting transition, whereas in pure poly(dA-dT)opoly(dA-dT) DNA force-induced strand separation seems still possible. In poly(dA-dC)opoly(dG-dT) DNA a high force transition is present, but no hysteresis between stretching and relaxing the molecules can be observed. Binding of the intercalator Ethidium Bromide disrupts the cooperativity of the BS-transition in a characteristic manner. At higher concentrations of the drug the force induced melting can no longer be observed. The minor groove binder berenil shows again different effects on the force spectra. In the low force regime the molecules become stiffer and the BS-transition loses cooperativity. At high concentrations the force at the end of the BS-transition is increased and mechanical strand separation is inhibited.

As all force spectra were characteristic for each drug, this technique might also become useful to investigate the interaction of other binding agents and drugs with DNA.

Nanometer scale characterization of silicon dioxide thin films with conducting atomic force microscopy (C-AFM) in ultra high vacuum

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The rapid decrease of structure size in modern semiconductor processing leads to the demand for new characterization techniques. The atomic force microscopy (AFM) and its already numerous derivatives give the possibility to determine certain characteristics on the nanometer scale both laterally and vertically. Here we apply conducting atomic force microscopy (C-AFM) to investigate thickness homogeneity and dielectric breakdown of very thin (4-8 nm) silicon gate oxides. To realize such measurements we use the ability of the OMICRON ultra high vacuum (UHV) AFM/STM (scanning tunneling microscope) to separately scan the surface in AFM mode and apply a voltage between the conductive tip and the silicon substrate at the same time. With this setup it is either possible to detect Fowler-Nordheim tunneling currents through dielectric films or to study dielectric breakdown effects.

To measure Fowler-Nordheim tunneling currents the voltage is ramped up to values below the determined breakdown voltage and the resulting current-voltage (I-V) curve is recorded. With our setup we can achieve a noise of the current signal in the range of 1 pA which gives us the possibility to detect very low tunneling currents. Because of the high sensitivity of the Fowler-Nordheim tunneling current to variations in the thickness of dielectric films this method is well suited to study the homogeneity of very thin gate oxides. Such homogeneity investigations are carried out by measuring I-V curves on a square grid covering an area up to $5 \times 5 \mu\text{m}^2$. The determined thickness variation of thermally grown gate oxides are in good agreement with the rms-roughness determined by conventional AFM measurements.

Further we use the C-AFM setup to cause dielectric breakdown of silicon gate oxides. From our experiments we obtain information about the distance dependence of consecutive measurements which is useful to draw conclusions about the charge distribution during electrical stressing. Additionally we can determine the gate oxide quality on a nanometer scale which is not possible with the macroscopic techniques commonly used in semiconductor industries. In these macroscopic measurements the dielectric breakdown is always caused at the weakest spot of the oxide.

Magnetic Induced Phase Transition in Two-Dimensional Colloidal Systems

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We study the phase behavior of a two-dimensional colloidal system under the additional influence of an external one-dimensional periodic potential. In our experiments we use a colloidal suspension of paramagnetic colloidal spheres ($\sigma = 4.5 \mu m$) which is applied to a glass substrate. In order to apply an external periodic potential, we evaporated thin lines of nickel onto the substrate and covered it afterwards with a protective poly(methyl-methacrylate) (PMMA) layer. When the periodicity of the nickel lines is chosen to be commensurate to the mean particle distance, we observe a magnetic induced phase transition from a disordered into an crystalline phase with hexagonal order (magnetic-induced-freezing, MIF). The phase behavior of the system is investigated by means of video microscopy as a function of the applied external magnetic field.

Mechanical Properties of Cells – AFM Studies

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The Atomic Force Microscope (AFM) has emerged into a valuable tool for cell biologists. The unique combination of operation under physiological conditions, the high accuracy in positioning a very small tip and the high sensitivity in measuring and applying forces allow the investigation of the mechanical properties of living cells with a lateral resolution of a few ten's of nanometers. The elastic properties of cells are determined mainly by the cytoskeleton, which is a cross-linked polymeric network. The elastic properties of the cytoskeleton can be probed by locally pressing onto the cell with a defined force and detecting the resulting indentation of the cell. By modelling the geometry and material properties of the sample and the tip, we can calculate the local elastic or Young's modulus, the basic material coefficient of elasticity. The analysis also yields the point where the tip first touches the sample. Thus maps of the unindented height of the cell and maps of the elastic properties of the cell can be obtained. This allows for instance the measurement of the local mechanical response of cells to various factors. Examples are the effect of drugs, different substrates or varying growth conditions. The possibility to study living cells with the AFM allows the investigation of dynamic cellular processes. One related question is the unclear mechanisms involved in the division of cells. The actin cytoskeleton and its dynamic reorganization presumably play an important role in this process, obviously its mechanical properties are involved here, however the actual mechanisms are not understood well. A persistent and important question is whether cortical relaxation outside of the equatorial region or increases in cortical contractility within the developing cleavage furrow alone drive cell division. The AFM was employed to track dynamic changes in the stiffness of the cortex of adherent, cultured cells along a single scan line during M phase from metaphase to cytokinesis. We were able to get first insights in the mechanical aspect of cell division. The equatorial region stiffens before any cleavage furrow morphology appears. This stiffening dramatically increases as the first indication of the furrow occurs. On the other hand, while occasional softening of cells at edges distal to the furrow occurs, this does not appear to be an obligatory event for cell division.

Noise analysis on the Light-addressable-Potentiometric Sensor (LAPS)

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A stable and reliable interconnection between living cells and electronic devices, that allows for the detection of extracellular potentials of single cells is the topic of several research projects. Most basic approaches like microelectrode or field-effect transistor arrays are based on individually contacted sensitive devices that are separated by insensitive areas. This implies the problem, that cells that grow on the interspace between the measurement sites cannot be monitored. A promising example for a potential sensor that overcomes this problem seems to be the light-addressable potentiometric sensor (LAPS). In contrast to the former examples it consists of a laterally completely unstructured EIS-structure (Electrolyte-Insulator (Si₃N₄, SiO₂) Semiconductor (Si)), where any point of the surface can be addressed by illumination with a pulsed light-pointer. The photo-generated charge carriers redistribute in the electric-field of the space-charge region at the insulator-semiconductor interface generating a photocurrent, that depends on the extension of the space-charge region and therefore on the surface potential of the illuminated area. By that means the working principle of the LAPS is equivalent to spatially resolved CV-measurements. For a successful use as a cell-sensor the LAPS has to fulfill several conditions. The spatial resolution, that is not only limited by the size of the light pointer but also by the lateral diffusion of the photogenerated charge-carriers, has to be in the range of the size of a single cell (10 μm). This has been achieved by the use of thin semiconductor substrates. In contrast to that the desired time and potential resolution (3 kHz, 10 μV) that are given by the characteristics of the cells action potential have not been reached, yet. In this study a fundamental experimental and theoretical noise analysis, based on the equivalent circuit of a MOS-capacitor, is performed. Mainly the total size of the device and the modulation frequency of the light-pointer are varied systematically. The results show, that besides a high stability of the light source (amplitude/noise > 10⁵) a remarkable reduction of the total size of the device to about 100 μm x 100 μm is required in order to reduce stray-capacitance and therefore noise in photocurrent sufficiently to permit the recording of typical cell-potentials.

Monte-Carlo-simulation of hard disks in a external periodic potential

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We examine a two-dimensional system of hard disks in a external sinusoidal potential. The system is simulated by a Monte-Carlo - method. We are interested in the phase diagram and the classification of the phase transitions, and especially in transitions like 'laser induced freezing' and 'reentrant melting'.

For the evaluation of the data we use finite size scaling methods, that allow us to extrapolate to systems of infinite size, and to learn something about the nature of the transitions. We compare our results with experiments, and test predictions of the Kosterlitz-Thouless theory.

The Influence of specific bounding properties in organic adsorbates to imaging and nanomanipulation with STM

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By comparing the results of our STM investigations on four different organic adsorbates (Adenin, Uracil, PTCDA, 8CB) we were able to reveal the influence of intra- and intermolecular bounding properties to the imaging process and nanomanipulation attempts.

The results are important for a better understanding of STM images and for optimizing the nanostructuring of organic adsorbates.

Changes in Adhesion Properties of Cells Studied on Hematopoietic Stem Cells

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Force spectroscopy with Atomic Force Microscope (AFM) has revealed various molecular properties like elasticity and unfolding forces of single molecules. In this work the strength of molecular bonds between stem cells are investigated.

With a single cell attached to the very end of a tip-less cantilever one can probe other cells and measure the force between adhesive molecules in the membrane of cells. By varying the duration and the area of the cell contacts, it is possible to investigate time dependent adhesion phenomena. As the pulling velocity plays an important role for the adhesion forces the dependence of the loading rate gives results for kinetics of the binding.

In this single cell investigation hematopoietic stem cells, which exist in parallel in two different states, were utilized. A very adhesive state when the cell is quiescent and the other when the cell rounds up and loose its adhesivity to the surface are optically distinguishable. This is the beginning of differentiation of the stem cell towards a blood cell.

The changes in the maximum force, the single molecular force and the tether length were probed. The maximum force that is built up between two non-adherent cells is for short contact times (1 sec), much higher (270 pN) than for an adherent cell and a non-adherent cell (110 pN). Although the single molecular forces (30 pN) do not show a difference. Also in the rupture length differences can be observed. Between two non-adherent cells the disconnection occurs at shorter distances than for the other cell pair.

Varying the contact time the maximum force increases up to 1 nN and the adhesion force of the last rupture from 30 pN at 1 sec to 80 pN at 1 min contact time.

It seems that for two cells in the same state the unspecific adhesion at the beginning of a contact is more pronounced but at longer contact times the specific adhesion between two different cells is developed.

Studies of the Energy Transfer in Single Phycoerythrocyanin Molecules

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Single molecule microscopy has successfully been employed to studies of energy transfer processes in photosynthetic pigments. Avoiding any ensemble averaging makes otherwise hidden data accessible. The power of this approach has recently been shown by Köhler and coworkers [1]. In this work we present studies of the energy transfer in single phycoerythrocyanin (PEC) molecules. PEC is part of the lightharvesting complex in cyano bacteria, absorbing in the green spectral region. The naturally occurring forms of PEC are trimers and hexamers. In these aggregates, the excitation energy is dissipated over all molecules before finally being transferred to the photosynthetic reaction center.

Knowledge of the monomeric subunits' photophysics is prerequisite for an understanding of the energy transfer process in PEC. The PEC monomer contains one absorber and two chemically identical emitters. Polarization modulated excitation and polarization sensitive detection allow us to determine the energy transfer rates between the chromophores in PEC. It is also possible to investigate the heterogeneity of the energy transfer coefficients of single PEC molecules and to determine the individual, static contributions to the bulk behavior. Furthermore, spectrally sensitive detection enables us to identify the chromophores responsible for photobleaching of single PEC molecules.

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