Nanoscience - linking disciplines

Joint Workshop

Venice International University
San Servolo
September, 27th - October 1st, 2004

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Ludwig-Maximilians-Universität

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VENICE
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Joint Workshop: "Nanoscience: linking disciplines", 27.09.-1.10.2004, Venice International University

Program:

Time	Monday 27-SEP-2004	Tuesday 28-SEP-2004	Wednesday 29-SEP-2004	Thursday 30-SEP-2004	Friday 1-OCT-2004
9:30	Opening	-	-	-	-
9:45	**Lars Samuelson** *Nanoscience education and Nanowire research @ Lund University*	**Andreas Zumbusch** *CARS-microscopy: High resolution microscopy without labels*	**Stefan Hell** *Fluorescence Nanoscopy through Reversible Optically Saturable Transitions*	**Vladimir Shalaev** *Plasmonic Nanophotonics*	**Patrick Cramer** *The multiprotein machine for gene transcription*
10:30	**Mark Welland** *Novel methods in nanostructuring*	**Klaus Ensslin** *Semiconductor nanostructures and quantum information processing*	***Alexander Högele*** *Spectroscopy of single self-assembled quantum dots*	**Bert Nickel** *Diffraction techniques to access (bio-)molecular ordering at hard interfaces*	**Xi Zhang** *From Interfacial Self-assembly to Macromolecular Nanostructures*
11:15	Coffeebreak	Coffeebreak	Coffeebreak	Coffeebreak	Coffeebreak
11:45	**Andreas Engel** *The Basel Nanoscale Science Program for Bachelor and Master Students*	**Khaled Karrai** *Laser cooling of microlevers*	**Jan von Delft** *Anderson-Excitons: the effect of a Fermi sea on emission and absorption spectra*	**Manfred Radmacher** *Investigation of cellular and molecular activity by AFM*	**Birgitta Whaley** *Rational design of optimally coherent solid state nanostructures*
12:30	Lunch	Lunch	Lunch	Lunch	Lunch
14:30	**Naomi Halas** *Nanoshells: New tools for manipulating light at the nanoscale*	**Peter Grütter** *Contacting nanoelectronic structures by SPM techniques*	Informal Discussions	**Christiane Ziegler** *Nanobioanalysis for medical applications*	
15:15	**Fritz Simmel** *Functional nucleic acids as components of DNA nanodevices*	**Hans-Andreas Engel** *Efficiency of general qubit measurements and readout of spin qubits via charge detection*		**Dieter Braun** *Interfacial Forces move DNA in Thermal Gradients*	
16:00	Coffeebreak	Coffeebreak		Coffeebreak	
16:30	**Wolfgang Parak** *Biological applications of colloidal nanoparticles*	**Roger S. Goody** *Studies of intracellular vesicular transport mechanisms*		**Erwin Frey** *Nanomechanics of Microfilaments*	
17:15	**Christoph Gerber** *A novel biosensing and diagnostic method based on nanomechanics*	**John Lupton** *Nanoscale Optoelectronics*		**Andrey Rogach** *Energy transfer with semiconductor nanocrystals*	
18:00	Welcome reception	Dinner		Dinner	
18:45		Poster Session I		Poster Session II	

Abstracts Talks

Interfacial Forces move DNA in Thermal Gradients

Dieter Braun

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Microfluidic separation techniques need a miniaturizable driving force. We use temperature gradients to move particles. Most molecules or particles at room temperature show a thermophobic behaviour with speeds of several µm/s for moderate heating. We heat with optical methods and used fluorescence techniques to measure temperature and particle concentration [1,2].

The effect is very pronounced for DNA. The origin of thermophoresis in liquids is not yet resolved, however experiments indicate that ionic particle-solvent interactions are crucial for thermophoresis of DNA [1,2]. We infer thermophoretic coefficients from microfluidic measurements of steady-state depletion profiles induced by infrared heating. Finite element calculations allow detailed simulation of thermophoresis in microfluidic settings. In thicker chambers of 50-1000µm height, convection together with thermophoresis yields an intricate and strong accumulation of DNA near the stagnation point of convection [1].

Interestingly, the same convective flow geometry can be used to exponentially amplify DNA with the Polymerase Chain Reaction (PCR) [3,4]. It is driven by the laminar flow which implements a natural temperature cycling between 65°C and 95°C. Such a convective PCR yields an unexpectedly fast and simple platform for diagnostics.

It is tempting to hypothesize based on [1-4] towards temperature gradient based molecular evolution of life far away from thermal equilibrium [5]. Porous rocks near hydrothermal vents or submarine seepages host steep temperature gradients. DNA or RNA could accumulate in pores by thermophoresis and replicate by temperature-cycling convection.

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The multiprotein machine for gene transcription

Patrick Cramer

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Protein-coding genes are transcribed by the central enzyme RNA polymerase II (Pol II). To elucidate the transcription mechanism, our laboratory uses X-ray crystallography to determine the three-dimensional molecular structure of Pol II in various functional complexes.
Structures of the ten-subunit core of yeast Pol II in free form (1, 2), in complex with DNA and RNA (3), and in complex with the inhibitor alpha-amanitin (4) have given insights into the mRNA transcription mechanism (reviewed in 5, 6). Based on this work two labs have independently determined a model for the complete 12-subunit Pol II last year (7, 8). We have subsequently solved the first structure of Pol II in complex with a transcription factor, the elongation factor TFIIS (9, 10). TFIIS stimulates a weak intrinsic RNA cleavage activity of pol II and is required for efficient mRNA proofreading and for escape from DNA arrest sites. A detailed model of this 13-polypeptide 536 kDa complex, derived at 3.8 Å resolution, shows a spectacular binding mode of TFIIS to the polymerase surface, and reveals that Pol II has a single tunable active site for both RNA polymerization and cleavage. After a summary of this work, I will present very recent structural and functional data that provide insights into the coupling of transcription to mRNA processing and into pol II recycling (11, 12).

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The Basel Nanoscale Science Program for Bachelor and Master Students

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In the Nanoscale Sciences physics, biology and chemistry overlap: these disciplines seek to understand structures that are smaller than 100 nm. In spite of this common interest there are interdisciplinary barriers that prevent optimal exchange of knowledge. Such barriers are among others related to particular vocabularies and different ways to approach and solve problems. To overcome these hurdles the University of Basel offers an interdisciplinary Bachelor curriculum in Science with Nanoscale Sciences as the major. A Master program is implemented as well. While in the second year of its existence about 50 students have enrolled in the Bachelor program, only a few Master students participate so far. The programs will be presented and our current experience discussed.

Efficiency of general qubit measurements and readout of spin qubits via charge detection

Hans Andreas Engel

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The readout of a qubit state is of central importance for quantum information processing. In special cases, the qubit state can be determined in a single measurement, referred to as single-shot readout. In general, however, the preparation and measurement need to be performed not only once but n times, where n depends on the qubit, the efficiency e of the measurement device, and on the tolerated inaccuracy (infidelity) alpha. In the first part of this presentation, I analyze such n-shot readouts for general qubit implementations and derive a lower bound on n in terms of e, which in turn provides a definition for the measurement efficiency e [1]. I then focus on spin-based qubits and GaAs quantum dots and analyze their n-shot readout based on a spin-charge conversion and charge measurement via a quantum point contact (QPC). Various implementations based on single or on double dots provide efficiencies e between 50% and 100%, allowing single-shot read out in the latter case. One can model the read out microscopically and derive its time dynamics in terms of a generalized master equation, calculate the QPC current and show that it allows spin read out under realistic conditions [1]. Recent experiments using a single quantum showed that single spin read-out is feasible with a high efficiency [2], where the Zeeman splitting on the dot allowed spin-to-charge conversion by coupling to a lead [3].

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Semiconductor nanostructures and quantum information processing

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Highly tunable electronic nanostructures are fabricated by AFM nanolithography. Transport experiments on coupled quantum structures will be presented in view of their potential as building blocks for a future quantum information processor. The focus will be on an electric field tunable singlet-triplet transition, the time-resolved detection of single electrons, and the local spectroscopy of the electronic properties of nanostructures using low-temperature scanning probe techniques.

A novel method for biosensing and diagnostics based on nanomechanics

Christoph Gerber

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Micro-fabricated silicon cantilevers arrays offer a novel label-free approach where ligand-receptor binding interactions occurring on the sensor generate nanomechanical signals like bending or a change in mass that is optically detected in-situ.
We report the detection of multiple unlabelled biomolecules simultaneously down to picomolar concentrations within minutes. Differential measurements including reference cantilevers on an array of eight sensors can sequence-specifically detect unlabelled DNA and is suitable for detection of specific gene fragments within a complete genome (gene fishing).
Ligand-receptor binding interactions, such as antigen recognition will be presented. Antibody activated cantilevers which bind to the indicator proteins – creatine kinase and myoglobin have been used. This technique enables several proteins to be monitored simultaneously and over time: detailed knowledge of the level of these proteins would allow life-saving treatments for patients suffering from heart attacks. Current methods for protein detection involve labelling of proteins radioactively - procedures that are time consuming and non-instantaneous. New styles of chemical cantilever activation as well as structuring of the cantilever surface will be explored to enable flexible deposition or coupling of a variety of receptor molecules for bio-/chemo-sensor application such as e.g. membrane protein recognition, micro-organism detection, enantiomeric separation. New coating procedures, enlargement of the active surface area by dendritic molecules as well as improvement of the receptor-cantilever chemical bond will be presented.
This underlying nano actuation mechanism has more wide-ranging implications. The forces involved ~1 nN, are sufficient to operate micromechanical valves and related fluidics devices which would also permit operation of micro and nanomechanical machinery. Since the transductions does not rely on external control systems delivery devices could be triggered directly by signals from single cell, gene expression, or immune responses.

Max-Planck-Institut für molekulare Physiologie

Abteilung für Physikalische Biochemie
Prof. Dr. Roger S. Goody

Studies of intracellular vesicular transport mechanisms

Roger S. Goody

Max-Planck-Institute for Molecular Physiology, Dortmund, Germany

Rab family GTPases, key regulators of membrane targeting and fusion, require the covalent attachment of geranylgeranyl lipids to their C-termini for their function. The enzyme catalyzing the post-translational prenylation reaction (Rab GGTase) differs from other protein prenyltransferases in that it requires a protein cofactor (rab escort protein, or REP), which also serves to keep the modified protein in a soluble form and to chaperone it to the appropriate membrane. Anotherprotein, GDI (GDP-dissociation inhibitor), shares these chaperoning properties. We have determined the structures of both prenylated and unprenylated Rab molecules as complexes with REP and GDI. The combined structural and kinetic information on these interactions provides explanations for the GDI effect and for the preferential interaction of REP/GDI with the GDP form of Rab proteins. It also provides a starting point for understanding targeting of the Rab proteins to specific membranes and throws further light onto mechanistic aspects of the prenylation reaction and the solubilizing properties of REP/GDI.

Contacting nanoelectronic structures by SPM techniques

Peter Grütter

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In this talk I will present three different areas of research pursued in my group. The common link is that we are investigating various systems with potential in nanoelectroncs by the technique of atomic force microscopy.
Atomically defined contacts to molecules are crucial if the electrical transport properties of molecules are to be understood. By comparing such atomically defined experiments to ab-initio modeling, a better understanding of the structure-property relationship is expected. Experimentally, we use a combined UHV AFM/STM/FIM (field ion microscope) that allows us to investigate the electromechanical properties of two terminal devices with atomically defined junctions.
The properties of self-assembled quantum dots can be measured by cryogenic electrostatic force microscopy. The addition of an extra electron manifests itself as a readily measurable increase of the electrostatic interaction between tip and sample. We will present initial results of InP quantum dots measured at 4.5 K.
Finally, we are using Magnetic Force Microscopy to investigate the potential of small, magnetostatically coupled ferromagnetic particles as cellular automata. The major issue is the switching field distribution of these microfabricated particles as a function of size, shape and processing conditions. This is investigated by MFM in the presence of external fields.

Nanoshells: New tools for manipulating light at the nanoscale

Naomi Halas

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Our abilities to create metallic nanoparticles and nanostructures of controlled sizes and shapes, to calculate and characterize their optical response and assemble and integrate them into new nanoscale optical architectures, has led to the emergence of a new field-Plasmonics. The design of nanoscale optical components is guided by the principle of "Plasmon Hybridization", a simple and intuitive picture which exploits an isomorphism between the mixing of electronic wavefunctions in molecular orbitals and the plasmon response of metallic nanostructures.

For the past few years our research has focused on the properties of a specific nanoscale plasmonic geometry called a nanoshell: a chemically synthesized concentric layered nanosphere consisting of a dielectric core surrounded by a thin, uniform metallic shell, developed within our research effort at Rice University. A nanoshell provides precise control over optical fields at subwavelength dimensions and can be considered a fundamental component of nanophotonics. With this core-shell geometry we have developed tunable resonant nanoparticles across the visible and much of the infrared region of the spectrum. We have recently shown that this core-shell geometry can be used to controllably and systematically optimize the surface enhanced Raman scattering (SERS) response of adsorbate molecules at the nanoshell surface, for chemical sensing applications.

Tuning the plasmon frequency of noble metal nanoparticles into the near infrared region of the optical spectrum, the "water window" of highest physiological transmissivity, gives rise to a wide range of important new biotechnological applications for these types of nanostructures. This approach has led to several new tools for the biomedicine, including a whole blood immunoassay that can measure physiological quantities of antigens of interest in five minutes with no sample preparation, optically triggerable drug delivery materials, and a highly localized, optically directed approach to cancer therapy.

Fluorescence Nanoscopy through Reversible Optically Saturable Transitions

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Since its discovery by Abbe in 1873, the diffraction barrier has received a lot of attention. However, the (nonlinear) subdiffraction microscopy concepts of the mid 20^{th} century remained either too vague or subject to unrealistic physical conditions. Consequently, until recently, all far-field fluorescence microscopes remained conceptually and practically diffraction-limited.

We discuss the principle of breaking the diffraction barrier through reversible saturable optical transitions. This principle was first proposed in the mid 1990's in the form of Stimulated Emission Depletion (STED) [1] and Ground State Depletion (GSD) microscopy [2, 3]. In all cases, the diffraction barrier is broken by a saturated depletion of the ground or the excited state of the fluorophore. The saturation level defines the size of the ultrasharp focal spot and the concomitantly enlarged bandwidth of the optical transfer function (OTF). We show that the resolution can be approximated by $\Delta x = \lambda/(\pi n) = \lambda/(\pi n)$, whereby I_{sat} is the characteristic intensity required for saturating the transition, and denotes the intensity applied [4]. Hence the quest for nanoscale resolution boils down to maximizing the saturation factor $\varsigma = I/I_{sat}$, which means increasing I, and if this is not possible, lowering I_{sat} [4-6].

We give first evidence of STED-microscopy displaying PSF of 10-20 nm FWHM, corresponding to a 15-fold enlargement of the OTF over Abbe's barrier. The success of STED stems from the fact that the saturation of the single-photon transition of stimulated emission provides strong nonlinearities at comparatively *low* intensities. The reason for that is simple but critical: Unlike in multiphoton events, the nonlinearity produced by saturation does *not* rely on the joint action of multiple photons, but stems from the population kinetics of the fluorophore states. Hence transitions that are easy to saturate (i.e. with low I_{sat}), allow huge ς at low intensities.

In consequence, in 1995 we have proposed as a further option to STED, the saturation of the triplet state [2], which reduces I_{sat} by ~ 10^3, and also the 'switching' between conformational fluorophore states [6-8], which gives another factor of 10^3. We have proposed that such saturable switches are encountered in photochromic compounds [7] and photoswitchable GFP-like proteins such as *asFP* [4, 6], which should render nanoscale resolution with the ultralow intensities of a lamp.

Finally, we show that a slightly modified version of our concept may serve as an alternative to the current X-ray and synchroton efforts in nanolithography [6] with the potential of providing material (nano)structures of any size and density with visible focused light.

References

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Spectroscopy of single self-assembled quantum dots

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Self-assembled quantum dots are semiconductor based structures with nanometer dimensions. Hence, the motion of charge carriers - electrons or holes - trapped in the potential of a quantum dot exhibits quantization in all three directions. Like in atom physics, discrete energy levels determine the optical spectrum of quantum dots. In contrast to atoms or molecules however, there's a high degree of tunability of quantum dots properties. Tailoring the sample design or modifying the solid state environment through electric and magnetic fields or strain allow for controlled manipulation. For example, the regime of a quantum dot can be switched in a controlled way from a quasi isolated two-level system to a system interacting with a two-dimensional electron gas simply by applying a dc voltage. Therefore, quantum dots represent an ideal model for the study of quantum mechanical properties of localized charged particles as well as their interactions with the Fermi-sea.
One of the basic experimental approaches to the examination of quantum dots is the optical spectroscopy technique. My presentation will highlight recent experimental results obtained by photoluminescence spectroscopy [1] and the newly developed absorption spectroscopy [2] performed on single self-assembled quantum dots.

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A. Högele *et al.*, Physica E **21**, 175 (2004);
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Laser cooling of microlevers

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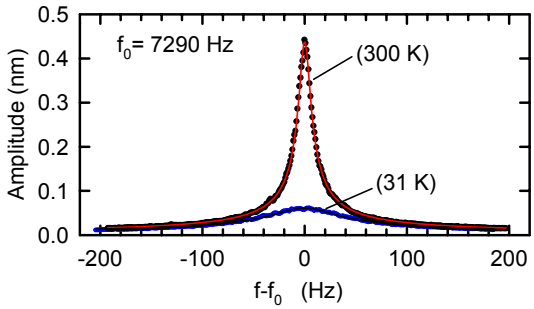
A silicon microlever forms one of the two mirrors of a Fabry-Pérot (FP) optical microcavity. When the cavity is tuned near one of its optical resonance, the density of stored photons is resonantly enhanced. Equilibrium is reached when the flux of stored photon equals the photon leakage, which is entirely determined by the mirrors transmission as well as the mirror separation. Since the lever is mechanically compliant, it moves under the effect of the resonantly increased photon-induced forces. So when subjected to a strong enough laser-field, the photon density stored in the Fabry-Pérot cavity changes because the lever mirror is pushed away. More importantly, during the lever motion, the photon leakage changes as well. Because this change is not instantaneous but delayed, it leads to a force acting on the lever that is proportional to the lever velocity. This corresponds to a damping force that leads to the laser cooling of the Brownian motion of the lever. As seen in the figure below, optical cooling was successfully obtained. We exploited systematically the analogies between the spectral properties of an atom and that of the optical Fabry-Pérot cavity. Laser cooling of an atom requires both a strong resonance in the optical spectrum, namely an atomic emission line, as well as a photon induced velocity dependent damping. The later condition is usually obtained by placing the atom in a field of interfering counter-propagating laser beams. Here, optical cooling was simulated in that i) the optical resonances of the micro-optical cavity mimics the atomic optical lines and ii) the light induced forces experienced by the lever damps its motion in a similar way radiation pressure acts on atoms.



Cavity cooling: Brownian noise amplitude spectrum measured in a frequency window centered on the lowest lever mechanical resonance at f_0 = 7290 Hz. The Fabry-Pérot cavity was slightly detuned from one of its optical transmission resonances. The larger peak in the noise spectrum data (dots) was obtained at low laser power (10 μW) and corresponds to the expected Brownian noise for 300K. The data in the smaller peak are obtained with a laser power increased to 200μW. The Brownian noise corresponds to 31 K.

Nanoscale Optoelectronics

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The electronic coupling of light and matter lies at the heart of optoelectronics. Although previously situated in the realm of the solid state, advances in microscopic and spectroscopic techniques mean that optical processes can now be studied at densities approaching the single photon limit. Device fabrication has traditionally evolved following top-down approaches subjected to ever decreasing scale sizes. In the long run, however, the avenue of bottom-up growth of optically active elements by employing naturally assembled structures conceptionally situated between the two extremes of single atoms and the solid state is more appealing. Novel optically active nanostructures include polymeric macromolecules and colloidal semiconductor nanocrystals, which both benefit from the richness and diversity of organic and inorganic synthetic chemistry.

Spectroscopic investigations of single colloids and macromolecules provide important insight into the fundamental electronic properties of the optically active unit. In bulk devices such as light-emitting diodes and solar cells, the electronic properties of the single molecule directly influence the overall device performance. We demonstrate that polymeric semiconductors, which can be thought of as long chains of carbon atoms, exhibit discrete colour centres on the single molecule level, which can interact with one-another through both coherent and incoherent energy transfer. Homogeneous spectral broadening is found to be independent of chemical structure, which in turn controls inhomogeneous broadening. Single molecule spectroscopy allows the identification of dynamic disorder, which plays an important role in describing the elementary photophysics. This can be accelerated by applying external stimuli such as thermal energy, enabling optical switching between different stable states of the molecule.

Semiconductor nanocrystals are traditionally described in the quasi-atomic picture, although their size typically exceeds that of even the larges macromolecules, and both electron-hole and electron-phonon correlations are of importance. As a large fraction of the atoms constituting nanocrystal quantum dots are situated at the surface, surface effects dominate the optical properties along with the obvious quantum confinement. The quantum confined Stark effect is particularly import in nanocrystals, as localised surface charges lead to a strong deformation of the effective electronic wavefunction overlap. By tuning the shape rather than the size of the nanocrystal from spherical to rod-like, we find that it is possible to employ the quantum confined Stark shift to gain insight into the microscopic dynamics of surface charges. In effect, the single particle spectroscopy becomes sensitive to charge transfer on the single electron level, posing an extremely effective sensor of the electrostatic environment. These observations are complemented by direct manipulation of the excited state using strong external electric fields, which reveal both linear and quadratic Stark shifts. Besides providing fundamental insight into the nature of excited state species in nanoscale emitters, controlling the excited state through external stimuli constitutes a direct demonstration of optical modulation on the ultimate size level.

Diffraction techniques to access (bio-)molecular ordering at hard interfaces

Bert Nickel

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We study molecular order and morphology of hard/soft interfaces. The variety of phenomena ranges from the determination of dislocation densities in crystalline organic thin films (pentacene) up to the nanostructure of lipid-cholesterol mixtures (rafts) in solid-supported membranes.

To access such information, new experimental approaches are developed using (high energy) synchrotron radiation (ESRF, HASYLAB) and high flux neutron sources (REFSANS at FRM II). In particular, our approach aims on the combination of microfluidics, diffraction and optical microscopy. This allows for the use of well proven preparation recipes, confocal microscopy and surface sensitive diffraction techniques (reflectivity, in-plane diffraction) on the same sample in a liquid environment.

Using this approach, materials science aspects such as response of functional biological interfaces to shear flow or the phenomenon of biomineralization could be studied as well at well defined conditions (molecular concentrations, flow conditions, temperature,...).

Colloidal nano-hybrid materials for biological labelling

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Recent advantages allow physicists to create smaller and smaller structures in a top-down approach, and chemists and biologists to assemble bigger and bigger structures in a bottom-up approach. Both approaches meet at the nanometre scale and it is possible to design interfaces between artificially created objects and biological molecules and organisms.

Materials can be designed with novel and tailored properties on the nanometre scale. Colloidal nanocrystals are one example of such efforts. By selecting the appropriate materials inorganic nanocrystals with fluorescent, magnetic, and other properties can be synthesized. The ultimate goal is to directly grow different materials on top of each other, and achieve in this way nano-objects with multiple functionalities. The creation of such materials is typically performed in organic solvents and thus these particles have to be rendered hydrophilic in order to make them water-soluble.

On a next level of complexity nano-building blocks can be assembled by the use of biological molecules. If one building block is functionalized with a ligand molecule, this building block can be linked to another building block that is functionalized with the appropriate receptor molecule. In order to create precisely designed assemblies of such biomolecule functionalized building blocks, the number of biomolecules per building block has to be exactly controlled. A biological linker molecule of particular interest is DNA, because of its programmability. The linkage of nano-building blocks with biological molecules is also reversible and can be controlled by external parameters as temperature and ion concentrations. Alternatively different building blocks can also be assembled by embedding them into the shell of polyelectrolyte polymer capsules.

By combining the synthetic approach to design multifunctional building blocks and by self-assembling them to controlled structure via linkage with biological molecules highly advanced objects can be created. They will have different functionalities, which in turn can be controlled by external parameters. An object of particular interest will be the creation of ligand-functionalized colloidal particles with fluorescent and magnetic properties.

These novel materials will be used for biological labelling and targeting purposes. Targeting is facilitated by directing the nano-composite structures due to their magnetic moment in magnetic field gradients and by ligand molecules immobilized on their surface. Labelling and the observation are facilitated by their fluorescent properties. In this way drug-targeting applications will be possible.

Investigation of cellular and molecular activity by AFM

Leif Riemenschneider, Stefan von Coelln, Markus Prass & Manfred Radmacher

Institut für Biophysik, Universität Bremen

We have used the AFM to follow biological processes under physiological conditions. At the cellular level, the AFM allows to measure local mechanical properties of the cytoskeleton and thus gives new insights in processes like cellular locomotion or cell division. The cytoskeleton consists mainly of filamentous Actin and Actin binding proteins. Actin binding proteins are responsible for the degree of cross-linking and the architecture of the network. For instance, the motor molecule Myosin will create internal tension in the network (so-called cortical tension). This could be observed during cell division in the cleavage furrow formed between the two daughter cells. When disabling Myosin biochemically with suitable drugs a softening of the cell will be observed.
On the molecular scale, we could demonstrate that the AFM allows following conformational changes of enzyme molecules during their activity. By watching fluctuations in the molecules height under different buffer conditions, e.g. in the presence of substrate or inhibitors, we could clearly detect the dynamics of single molecules by AFM. Recently, we have started employing single enzyme molecules, which are immobilized to the very apex of an AFM tip, to modify locally suitable samples. Thus we have demonstrated for the first time a new surface modification technique, which we called enzyme assisted nano-lithography.

Funneling and Recycling of Excitons in Semiconductor Nanocrystal Assemblies

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In semiconductor nanocrystals the electronic energy gap is determined not only by the material but also by the size of the nanocrystals. This allows constructing an energy gap gradient normal to multiple layers of nanocrystals whereby the diameters of the nanocrystals are monotonically increasing or decreasing in subsequent layers. For fabrication of such structures, the layer-by-layer technique which is based on alternating adsorption of oppositely charged species can be applied. We observe a highly efficient funneling of excitation energy from layers comprising smaller nanocrystals towards the layer with the largest nanocrystals. Most important, not only excitons in radiative states are transferred but also excitons from trapped states, usually lost for luminescence, can be effectively recycled, hence increasing the overall luminescence yield.

Nanoscience education and Nanowire research @ Lund University

Lars Samuelson

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Since 16 years, when the Nanometer Structure Consortium was formed, Nanoscience has been a major field of research at Lund university. A few years ago it was recognized that it would be of great value to get an educational program in the field started in Lund, resulting in a complete 5-year University education, which was started in 2003, called "Engineering Nanoscience" [1].
From about the year 2000 our research has had an increasing focus on Semiconductor Nanowires, with the research focused on aspects of nanowire growth and related technologies, advanced characterization, physical properties and different applications of nanowires in electronics, photonics as well as in life-sciences. This will be the primary area of my presentation [2–15].

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Plasmonic Nanophotonics

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There is ample evidence that photonic devices can be reduced to the nanoscale using optical phenomena in the near field, but there is also an incompatibility between light wavelength at the *microscale* and devices and processes at the *nanoscale* which must first be addressed. Plasmonic nanostructures can act as nanoantennae and thus serve as optical couplers across the nano–micro interface. Recent advances in this rapidly developing area now enable us to mount a systematic approach toward the goal of full systems-level integration of photonics with nanotechnology using nanoscale plasmonics. Plasmonic nanophotonics also promises to create entirely new prospects for guiding light on the nanoscale, some of which may have revolutionary impact on present-day optical technologies. In this talk I outline some of our recent studies on manipulating light and sensing molecules with plasmonic nanostructures.

Functional nucleic acids as components of DNA nanodevices

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DNA and RNA can be used to build supramolecular structures and devices by self-assembly. However, the design possibilities for DNA nanostructures are limited when only simple base-pairing interactions are considered. Furthermore, to add functionality to DNA-based nanostructures, DNA usually has to be modified chemically. One of the possibilities to increase the versatility of DNA nanoconstruction is to utilize non-standard base-pairing interactions. In contrast to the formation of a double-helix as in the Watson-Crick base-pairing scheme, non-standard interactions can give rise to unusual three-dimensional folds. In some cases, such non-standard structures have catalytic properties (ribozymes) or can bind specifically and strongly to proteins or other molecules (so-called aptamers). In fact, these properties can be evolved in selection experiments on random pools of DNA or RNA sequences. We will here give an overview of work by our group and other groups in which attempts are made to utilize the binding and catalytic properties of DNA/RNA aptamers and (deoxy)ribozymes for nanoscale construction. With such structures, molecules can be bound, free-running machine-like devices can be devised and simple computations can be performed.

Anderson-Excitons: the effect of a Fermi sea on emission and absorption spectra

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Recent experiments measuring the emission of exciton recombination in a self-organized single InGaAs quantum dot (QD) have revealed that novel effects occur when the wetting layer surrounding the quantum dot becomes filled with electrons, because the resulting Fermi sea can hybridize with the local electron levels on the dot.
Motivated by these experiments, we study an extended Anderson model, which describes a local conduction band level coupled to a Fermi sea, but also takes account of a local valence band level.
We are interested, in particular, on how many-body correlations resulting from the presence of the Fermi sea affect the emission and absorption spectra.
Using Wilson's Numerical Renormalization Group, we calculate the zero-temperature emission (absorption) spectrum of a QD which is finally (initially) in the strongly correlated Kondo ground state.
We predict two features: Firstly, the threshold energy ω_0 - below which no photon is emitted (absorbed) - shows a marked, non-monotonic shift as a function of the exciton binding energy U_{exc}.
Secondly, we find that the spectrum diverges at ω_0, in close analogy to the well-known X-ray edge absorption spectrum.

Vapour phase growth of Silicon Carbide and Zinc Oxide based nanostructures

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Several techniques have already been developed for synthesising silicon carbide (SiC) material in the form of nanospheres and nanowires/rods. Here, we report the synthesis of a distinctly different kind of SiC nanostructure in the form of three-dimensional crystalline nanowire based flower-like structures and arrays of coaxial rods. Interest in such structures centres around the combination of a simple growth process based on SiC nanowire formation, with a resultant complex structure having potentially complex mechanical and optical properties; the latter a consequence of the wide band gap of bulk SiC. The synthesis of these SiC nanostructures is via a vapour-liquid-solid (VLS) process, on which a detailed study of both the chemical and structural composition has been carried out. In addition, we demonstrate the unique physical properties of the films through a photoluminscent study of the optical properties and a dynamic wetting study of the surface chemical/mechanical properties.

Rational design of optimally coherent solid state nanostructures

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Nanostructured materials present new opportunities for spintronics and for coherent applications such as quantum information processing. In order to make progress in these areas, new materials optimized for spin transfer are required. I shall present theoretical studies of electronic structure and transport in coupled nanostructures such as metal and semiconductor clusters linked by molecular bridges, that reveal how spin transport may be enhanced by suitable molecular design.
Applications to experiment will be given where relevant.

From Interfacial Self-assembly to Macromolecular Nanostructures

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Macromolecular chemistry is not limited anymore to repetitive improvement of already successful mass polymers, and it has expanded more vividly into the fields of functional materials, nanotechnology, surface chemistry, supramolecular systems, life science and biomedicine. We have been interested in rational design and synthesis of self-organizing building blocks for the construction of nanostructured organic thin films with tailored functions. In this presentation, two methods for fabricating macromolecular nanostructures through interfacial self-organization will be discussed. On the one hand, we have fabricated polyelectrolyte multilayer by layer-by-layer (LbL) alternating deposition of poly (diallyldimethylammonoium chloride) and poly (4-stylene sulfonate), and demonstrated that the multilayer can be used as a good matrix for electrochemical deposition, leading to formation of inorganic nanomaterials with controlled morphology. More important, this research provides a new way for the fabrication of super-hydrophobic properties, by combination of LbL technique and electrochemical deposition. On the other hand, we have established a unique and simple method of stabilizing surface micelles of a low molecular weight surfactant, 11-acryloyloxyundecyltriehtylammonium bromide, through in situ intramicellar polymerization initiated by gamma ray irradiation. The robust polymerized surface micelles are important not only for better understanding the self-assembly of surfactant molecules in colloid and interface, but also for further application in templating synthesis of nanomaterials.

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Nanobioanalysis for Medical Applications

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In the first part, force measurements and scanning force microscopy results in dental clinical research are presented. Biofilm formation becomes more and more important in technical as well as in medical applications. As an example, the adsorption of proteins and other macromolecules from the saliva on enamel, the hard, outermost surface of a tooth, leads to the formation of a biopolymer layer called pellicle. The pellicle layer protects the tooth against acid attack and lowers friction between the tooth and other teeth or other hard materials. It also offers a remineralization depot for Ca apatite, out of which most of the tooth is composed. However, bacteria, including caries bacteria, may also attach to the pellicle which then forms a complex layer called plaque. The understanding of pellicle formation hence presents an important key of preventive dentistry and caries research.
Here, we present scanning force microsocpy and spectroscopy measurements which allow the determination of adhesion forces between different saliva proteins and substrate surfaces used in dentistry. Conformationally stable molecules such as bovine serum albumin (BSA) show only slight differences in their adhesion force if compared to conformationally more flexible proteins such as lysozyme. Furthermore the morphology of in-vivo formed pellicle layers on these substrate surfaces could be analyzed.

In the second part, cantilever-based sensors are presented which will be used for medical applications. Here we present the technical realization for cantilever-sensors which work in-situ in the liquid phase.

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CARS-microscopy: High resolution microscopy without labels

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During the last decade, different techniques of non-linear optical microscopy have become versatile tools for cell microscopy. The latest addition to this list is Coherent Anti-Stokes Raman Scattering (CARS) microsopy [1]. Compared to other non-linear mircroscopies, CARS microscopy has the advantage of offering chemical sensitivity, i.e. the presence of vibrational bands is monitored directly and no external labeling is necessary. Therefore, microscopical investigations can be performed under the most benign experimental conditions.

Three-dimensional CARS microscopy has first been demonstrated with a pulsed fs-laser system. Recent years have seen many different technical developments. On one hand, new techniques such as CARS correlation spectroscopy have been explored [2]. On the other hand, many approaches were aimed at improving the signal to background ratio by reducing and suppressing the non-resonant signal. After a general introduction to CARS microscopy, we will demonstrate a new simple and straightforward approach, how this goal can be achieved. It will be shown how the bandwidth of broad-band fs-laser pulses can be spectrally focussed onto the vibrational resonance under investigation. For this purpose, the excitation pulses are chirped in order to keep their instantaneous frequency difference constant. Spectral features that are 100 times narrower than the excitation bandwidth are easily resolved. This approach is generaly applicable for other non-linear microscopical techniques. Its application to microscopy will be demonstrated with imaging of various biological samples.

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Abstracts Posters

Energy transfer excitation of single molecules using colloidal semiconductor nanoantennae

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We present experiments on the energy transfer in a hybrid system of single semiconductor core-shell nanoparticles embedded in a film of dye molecules. Hybrid systems based on inorganic and organic materials have been shown to offer advantages over purely organic or inorganic systems, namely the large absorption cross-section and the high photostability of nanoparticles combined with the high quantum efficiency of dyes. This results, for example, in a high overall photoluminescence efficiency due to energy transfer. Here, a single-molecule approach has been taken, showing that nanoparticles can act as tiny antennae collecting the excitation energy and delivering it to surrounding dye-molecules via energy transfer on a length scale of some nanometres.
Thanks to a careful selection of the optical absorption of the nanoparticles and the dye, the nanoparticles can be optically excited in an efficient way, whereas the excitation of the dye by direct absorption of the laser light is negligible. Furthermore, the dye's absorption has a large spectral overlap with the photoluminescence of the nanoparticles, making energy transfer possible. When illuminating the sample, a local increase of the dye's photoluminescence at the spatial positions of the individual nanoparticles is observed, demonstrating energy transfer from the nanoparticles to the surrounding dye molecules. In addition, a pronounced quenching of the nanoparticles photoluminescence is visible, which is strongly dependent on the concentration of dye molecules in the film and the resulting distance between nanoparticles and dye molecules. Accordingly, the lifetime of the nanoparticle photoluminescence is shown to decrease when embedding them into a film of dye molecules. Time-traces of the photoluminescence of single dye-molecules excited via energy transfer exhibit the typical blinking behaviour known from single quantum emitters, proving the observation of energy transfer on a single-particle level. Finally, irreversible one-step photobleaching of the dye acceptor leads to a recovery of the donor emission. Energy transfer excitation of dye molecules provides an unique way of addressing single emitters in an ensemble using spatially discrete nanoantennae.

Two-dimensional DNA-nanostructures

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In this project, the base-pairing interactions of DNA are utilized for the construction of complex DNA networks. Two molecules of DNA will bind to each other if their base sequences are exactly complementary – they will not bind if they are not.
Based on this "simple" rule we designed basic motifs such as linear structures, four-arm junctions and tiling lattices which form the building blocks of more complex molecular networks. These constitute the basis of DNA-based supramolecular structures which can be used for the arrangement of functional nanoscale components such as nanoparticles or quantum dots and even for the assembly of nanoelectronic circuits.

Photoconductivity of composite films of CdSe nanocrystals and C60 molecules

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The electrical properties of semiconductor nanocrystals (colloidal quantum dots) are of growing interest for optoelectronics. Due to their size-dependent optical band gap, they are promising materials for many applications, e.g. for photodetectors. Here we present the results from wavelength dependent photoconductivity measurements on composite films comprising CdSe nanocrystals and C60 molecules.
Composite films were obtained by dropping toluene solutions of CdSe nanocrystals and C60 molecules, either bare or mixed in different proportions, on a pre-structured substrate with a subsequent drying in air. Substrates were prepared by evaporation of two finger-like gold electrodes with a 5 μm or 10 μm spacing in between on standard glass plates using a single optical lithography step. Photocurrent measurements were done using a parameter analyser; wavelength dependent photoconductivity measurements in the range of 400 nm to 800 nm were performed using a halogen lamp as an excitation source and a monochromator.
We show a highly increased photoconductance of the composite CdSe/C60 film compared to the bare CdSe and C60 films, which reached more than three orders of magnitude. On the other hand, dark conductance of the composite film increased much less. A clear correlation was found for the wavelength resolved photoconductance and optical absorption spectra of the composite films, with positions of the maxima in the both spectra being dependent on size of CdSe nanocrystals. The increase of the conductivity of the composite films under illumination is explained by the creation of free charge carriers due to the separation of electrons and holes under optical excitation, with C60 molecules acting as electron acceptor.

Terahertz Microscopy for Nanodevices

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One of the most crucial parameters for quantum electronic devices is the electronic dephasing time. However, the measurement of dephasing in a quantum dot is challenging because electronic contacts can influence significantly the dynamic evolution of the quantum system. For this reason a contact-free microscopic probe of the dephasing process is required. In this contribution we will present a novel THz technique that fulfills this requirement and has the potential for imaging electronic dynamics with submicron resolution.

Many works have shown that THz waves are an excellent probe for monitoring charge carrier dynamics in the time domain. However, the long wavelength of THz radiation (1 THz corresponds to 300 □m) limits the spatial resolution that can be achieved by applying classical microscopic techniques. Recently, we have demonstrated an apertureless THz scanning near-field microscope (THz-SNOM) where the dielectric permittivity of a surface is sampled by a metallic tip. Resolutions down to 150 nm were achieved giving THz spectroscopy access to the nanoworld [1]. The sensitivity of the THz-SNOM on charge carrier distributions was investigated in experiments on n-doped GaAs structures. Electromodulation of the surface depletion region showed that THz microscopy can image the dielectric properties of electrons that are confined in an area of about 1□m2.

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Simultaneous mapping of two affinity-different binding sites of DNA-LexA repressor protein from Escherichia coli using the AFM

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The precise mapping of protein-binding sites on DNA and the determination of the DNA binding sites in different cellular states are vital for understanding complex cellular functions and mechanisms, in particular the control of gene expression. The repressor protein LexA is known from classical Biophysical and Biochemical data to control the expression of 31 genes in E. coli. Although LexA binding sites from those genes have similar sequences they are not identical, which indicates that LexA interacts with different affinities to these sites. Here, we describe an AFM-based approach for mapping two DNA binding sites simultaneously and revealing the different affinities of complex formation between LexA protein and DNA. This approach provides a rapid and efficient way of creating a large-scale physical DNA map with singular or duo protein binding sites on a gene. The protein binding sites were uniquely mapped by distinguishing two termini of a constructed linear DNA fragment. In addition by showing the binding of protein LexA to both the DNA loci of a gene, we are able to delineate a higher frequency of promoter occupancy at the recA site, in comparison with the operator yebG, by a single LexA repressor molecule; thus differentiating the binding affinity of the protein along a gene.

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Simple method to isolate individual polymer chains on substrate for direct measurement of the desorption force

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Single-molecule force spectroscopy (SMFS), based on atomic force microscopy (AFM), has become a versatile platform for studying intermolecular and intramolecular interactions with its extremely high force sensitivity.[1-2] The preparation of a “single-molecule” sample is a key issue in SMFS. Improper prepared samples often lead to a result of multiple molecules. Herein we present a method to prepare a single-molecule sample.[3] By silanization, the hydroxyl groups’ tailored surface can be covered by amino groups; however, the dispersed hydroxyl groups’ tailoring area remains because of the imperfection of the modification. These surface defects enable us to isolate polymer chains individually on the substrate. The SMFS measurements provide proof that polyelectrolyte chains are individually isolated by the unfavorable walls. Moreover, by controlling the time of silanization, we can adjust the amount of adsorbed polymer. There is a correlation between the force pattern and the adsorption conformation of the polymer. The long plateau in this case reflects a flat conformation of polymer chain at the interface. This method would be significant for single-molecule chemistry and physics because of the high performance of single-molecule isolation and the ease of the preparing process as well as the potential for automatization of single-molecule detection.

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Neutron diffraction on biofunctional interfaces

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Linking biological molecules to solid interfaces is in the heart of biotechnology. Examples of such hard (metalls, semicondictors, ceramics etc.) and soft (polymers, membranes, proteins etc.) matter interfaces are applications such as implants, bio-sensors, DNA-chips, and cell-semiconductor systems. In order to confine biological molecules to interfaces, nature uses chemical forces and self-assembly principles, allowing to assemble functional units to larger assemblies. Men made (biomimetic) systems such as templated membranes try to imitate biological function using the same self-assembly principles. Thus, a molecular picture of self organization processes at interfaces is a keystone for building up such systems. As we shall demonstrate, recent instrumental developments at the FRM2 neutron source may allow to access such structural aspects of biofunctional interfaces in great detail.

Exciton recycling in graded gap nanocrystal structures

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The energy of the excitonic transition in CdTe nanocrystals (NCs) can easily be tuned over the whole visible spectral range by changing their size. NCs can also be used as building blocks for mesoscale architectures, often called artificial solids. One example is a set of adjacent layers of differently sized CdTe NCs that shows high energy transfer rates [1].
Motivated by natural antenna complexes and by semiconductor heterostructure lasers, we present a cascaded energy transfer (CET) structure made of CdTe NCs. Funnel like band gap profiles are realized applying layer-by-layer assembly to CdTe nanocrystals of distinct sizes.
Directed energy transfer between layers of differently sized NCs forces the excitation energy into the active zone, i.e. into a single layer of the largest NCs placed in the center of the funnel. Photoluminescence (PL) and photoluminescence excitation (PLE) spectra show two important features. First, there is negligible emission from the smaller sized NCs. The small particles apparently serve as donors and the excitation energy is efficiently transferred along the gradient towards the layer of the largest particles. Second there is a clear enhancement of the emission intensity of the largest NCs. The CET structure with a single emitting layer shows a 4 fold increased quantum efficiency compared to a reference sample containing 7 layers of the largest NCs and therefore an increase in the final exciton density in the emitting layer by a factor of 28. This super-efficient exciton funneling can be explained by a recycling of surface trapped excitons that are usually lost for photoluminescence [2].

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Conductive Cantilevers for AFM on Biological Samples

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The atomic force microscope has proven to be a powerful tool to image proteins with sub-nanometer lateral resolution under near-native conditions, i.e. at room temperature, in buffer solution and at ambient pressure. In addition, the high signal-to-noise ratio enables the probing of single proteins, rather than the average configuration of large ensembles of proteins. As a result, proteins have been observed in different conformations.
In this project we aim to have local electrical access to the sample in the AFM, while obtaining topographic information at normal resolution. To this end, we developed conductive cantilevers. With such cantilevers it should be possible to alter the conformation of proteins (e.g. ion channels), as function of the applied voltage, or to detect local charge transport through a protein (e.g. photosystems). In this poster the current status of the cantilever development is presented.

Dynamics of Excitons in Coupled Quantum Well Structures

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The experiments to learn more about motion and interaction of long-living excitons in coupled quantum wells (QW) [1,2] are carried out in semiconductor heterostructures at low temperatures (<4 K). These epitaxially grown samples contain two GaAs-QWs separated by a thin Al0,3Ga0,7As tunnel barrier. Using photo and electron beam lithography, nano-patterned gate structures are applied to these samples allowing external control over the sample's band structure.

Creating long-living excitons
Excitons in bulk semiconductors exhibit short life times of typically only few nanoseconds. By applying an electrial field perpendicular to the QW layer of our heterostructure, photo-generated excitons perform a transition to spatially indirect excitons. This means their electrons and holes are located in different QW layers. The thin tunnel barrier in between prevents instantaneous recombination, and the excitonic life time increases by orders of magnitude [1,2].

Moving long-living excitons
The long life time of indirect excitons allows studying their drift properties. In order to apply external force on neutral excitons, we create an excitonic potential landscape by superimposing an additional lateral modulation on the vertical electrical field (quantum confined Stark effect) [3,4]. Excitons can be driven in-plane by variation of the excitonic potential landscape in-time.

Trapping long-living excitons
In order to to experimentally investigate high excitonic densities at low temperatures, this project also aims to combine both driving and confining excitons (excitonic trap). In first experiments, we observed excitonic confinement in a structure providing SiO2-dots on its surface: excitons were trapped in ring-shaped regions surrounding the dots.

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Nanotribology of alkali halide surfaces

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Due to their simple structure and the weak interaction with the probing tip, ionic crystals are ideal systems for studying nanoscale phenomena by atomic force microscopy in ultra-high vacuum. With the present contribution we discuss fundamental friction and wear processes observed on KBr and NaCl. At extremely low loads (< 1 nN) a "superlubricated" regime of motion can be achieved, in which the probing tip alternately follows or precedes the cantilever driving it without energy dissipation. At higher loads sliding becomes discontinuous. At room temperature the occurrence of discontinuities is modified by thermal vibrations, which lead to a logarithmic velocity dependence of friction. The elastic regime is easily overcome on KBr. In such a case the tip starts to pick up material from the surface and to redeposit it in mounds which reproduce the structure of the primeval surface. On larger scales, ripple patterns are formed perpendicular to the scan direction, with wavelengths of the same order of the tip radius of curvature.

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 Bucky-Balls 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 0.7nm 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 1nm 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. www.nano-science.de/stm

Aminoterminated Surface Patterns for Guided Assembly of Nanoparticles

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The fabrication of nanostructures is recently attracting considerable interest. We focus here on the nanolithographic modification of organic self-assembly monolayers of n-octadecyltrichlorosilane (OTS) on silicon. Nanopatterns of –COOH functions are formed by the local electrochemical oxidation of surface terminated –CH3 groups of the OTS[1,2]. By subsequent self-assembly of a short, commerially available silane, as provided by aminopropyltrimethoxysilane (APTMS), the top –COOH groups are transfered into aminoterminated surface patterns. These templates are promissing candidates for the site-selective assembly of e.g. negatively charged nanoparticles via electrostatic interaction.

Here we present studies on the stepwise self-assembly of the APTMS molecules and the subsequent assembly of negatively charged silica particles. Preparation conditions are optimized on large surface areas and are finally applied in lithographically confined surface areas.

Additonal derivatization routines for –NH2 terminated templates are envisioned for many other nanomaterials, i.e. carbon nanotubes, etc.

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[2] R. Maoz, E. Frydman, S. R. Cohen, J, Sagiv, Adv. Mater. 2000, 12, 725.
Single Molecule Spectroscopy of Cy-5 dyes in MCM-41 particles

Single Molecule Spectroscopy of Cy-5 dyes in MCM-41 particles

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The synthesis and the application of 'host-guest' materials, which are binary materials composed of a molecular guest species such as organic dye molecules incorporated into a solid host matrix such as nanostructured porous silica, has reached a high level of sophistication. Single molecule spectroscopy poses a powerful tool to characterise and to address these materials.

The spectral properties of a host-guest material are be analysed with room temperature confocal microscopy. In the presented study we obtain emmission spectra from individual Cy-5 molecules covalently attached to the inner surface of a MCM-41 host material. The mesoporous material, which has cylindrical pores of 3 nm diameter, is flooded with different solvents. The static and dynamic fluorescence properties of the individual molecules, like intensity and the wavelength of the fluorescence maximum, react to molecules' surrounding. It is shown that the spectra of the individual Cy-5 molecules are affected by the polarity of the solvents, thus revealing the presence of solvent in the immediate vicinity of the molecule. The Cy-5 molecule, in this case, acts like a sensor in a nanometre sized pipe.

Monitoring Conformational Diversity in Self-Organized Monolayers with a Scanning Tunneling Microscope

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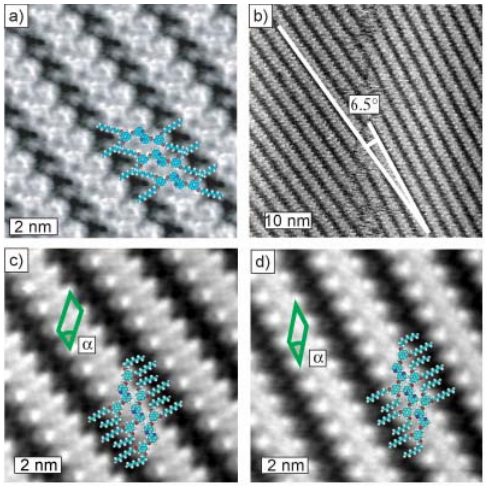
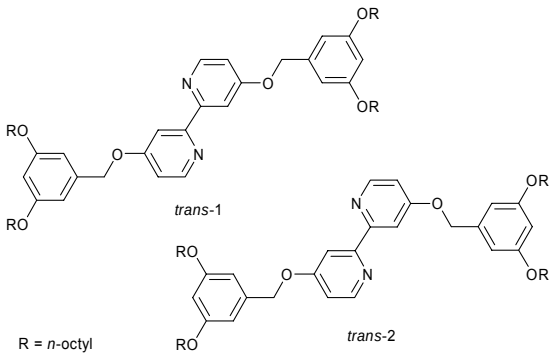
The direct imaging of chemical species at molecular and submolecular levels allows to probe self-organized structures using scanning tunneling microscopy (STM). The evaporation of solutions of 2,2'-bipyridine-dendrons on a graphite surface gives such highly ordered monolayers in multiple conformations on a graphite surface. The near atomic resolution allows us to assign two conformers, which spontaneously and rapidly form molecular domains under ambient conditions. Within a molecular domain, only one conformer is present and domains of different conformers are observed side by side. No preference for one conformer was observed. In the two domains in Fig. b, the conformations trans-1 and trans-2 (Scheme) can be assigned by comparing the relative positions of the benzyl groups of two neighboring molecules. Additionally, conformer trans-2 (Scheme) occurred in two different packing arrangements (Figs. a and c). The STM imaging at near atomic resolution allows a detailed conformational analysis to be made of this dendrons at room temperature and in air.





Manipulation of Single DNA Molecules on a Molecular Workbench

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Surfaces are processed to serve as a molecular workbench for bioparticles. Fluid cationic lipid layers are prepared on glass or plastic surfaces to force DNA molecules to adsorb onto the lipid plane while allowing the DNA molecule to diffuse freely in the lipid plane. We found that on nanostructured surfaces long DNA threads (50 000 bps) will orient along one direction and form a straight linear molecule. A theory has been developed to explain this phenenomen. In the future, the straightened and flat conformation of the DNA will allow the study of the dynamic interaction with single proteins by fluorescence microscopy. Furthermore, T5 phages have been found to attach to a hydrophobic plastic surface. A constant flow is applied and the protein FhuA which triggers the ejection of the phage DNA is added. The ejection process can now be monitored dynamically in vitro with fluorescence microscopy. Surprisingly, it is found that the ejection takes place in steps and ends with only about 60% of the full DNA length ejected. An explanation of this lays in the native nicks - single stranded interruptions of the otherwise double stranded DNA - of the T5 phage.

Optical Cooling of Micromechanical Systems

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A gold coated Si-cantilever forms one mirror of a Fabry-Pérot micro cavity. The flexible cantilever is subjected to photon-induced forces which are dependent on the mirror separation. A slight change in mirror position of the thermally vibrating lever will cause the light intensity and consequently the force on the cantilever to adapt to the new situation after a characteristic delay time. Because of this time lag the photon-induced force is dependent on the lever velocity and thus acts like a cold damping force. The significant difference to passive damping forces is that the described mechanism does not alter the thermal excitation of the lever, which still obeys the fluctuation-dissipation theorem.This allows to control the effective temperature of the lever's vibrational mode.
Using a monochromatic laser beam we can obtain not only a shift in the resonance frequency but a dramatic change in linewidth, which can be interpreted as an effective cooling or heating of the lever vibration. When the cavity is slightly detuned from resonance by decreasing the separation between the mirrors, the thermal motion of the cantilever gets self-amplified or alternatively increasing leads to cooling. The behavior can be understood and modelled by the action of the photon gas in the system. The cooling efficiency depends on the intensity of the laser beam, the mechanical Q-factor and the finesse of the cavity.

Magnetic Nanostructures – Fabrication via the Derivatization of Chemically Active Surface Templates and Characterization with MFM

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The technological significance of magnetic data storage has been tremendously increased in the last decades and a high amount of economical benefit originates in the rapid development of fast and large storage devices.
Pushing the size limits of individual storage bits to a miniumum is therefore a key issue for fundamental and applied research. The exploration of the magnetic properties, their dependence on the size and shape of individual storage device structures is an important task that can be investigated conveniently by means of Magnetic Force Microscopy (MFM).
We demonstrate a new lithographic approach to generate magnetic nanostructures directly on predefined chemically active surface templates. Therefore a self-assembly monolayer of n-octadecyltrichlorosilane (OTS) molecules is employed as a substrate which can be locally chemically activated by means of a site-selective oxidation, which is mediated by a conductive Scanning Force Microscopy (SFM) tip.[1] By application of a negative tip bias voltage the surface terminated –CH3 groups are electrochemically converted into reactive acid groups that are capable for further derivatization routines,[2] such as to generate magnetic nanostructures of arbritary shape and size. The fabrication of these structures is demonstrated and MFM investigations elucidate the magnetic properties of the nanodimensional structures.

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[2] S. Hoeppener, R. Maoz, S.R. Cohen, L.F. Chi, H. Fuchs, J. Sagiv, Adv. Mater. 2002, 14, 1036.

Absorption spectroscopy of single self-assembled quantum dots

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Interband absorption into excitonic states of a single self-assembled quantum dot has been recently reported [1,2]. Optical transitions are observed directly as a reduction in transmission signal when the exciton energy is tuned into resonance with a narrow band laser. With this technique, the ground state of neutral excitons confined in a single quantum dot is probed as a function of electric and magnetic field at low temperature. We present experimental results which profit from all the advantages of laser spectroscopy, namely high spectral resolution, line shape determination and direct access to the oscillator strength. The linewidth of the exciton ground state was found to be broadened due to spectral fluctuations. Nevertheless, the experimental data obtained from saturation spectroscopy of the corresponding transition are well modelled as a two-level system.

We also investigated the charge dependence of the electron-hole exchange interaction in a single quantum dot. Transmission spectra of the neutral and the singly charged exciton are Lorentzian-shaped lines with a linewidth of ~ 2μeV. The neutral exciton transition reveals two linearly polarized resonances separated by the fine structure splitting. The splitting is typically 15-40μeV and arises through electron-hole spin interaction within a dot with an anisotropic in-plane potential. However, when the quantum dot is filled with one additional electron, the resonant absorption of the singly charged exciton shows only one line. The total electron spin is now zero so that the electron-hole exchange vanishes. This demonstrates how the spin mediated interaction can be turned off simply by applying a small dc voltage.

[1] B. Alèn et al., Appl. Phys. Lett. 83, 2235 (2003).
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Low noise and fast scanning force microscopy with small cantilevers

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In scanning force microscopy and in force spectroscopy, the measurement speed and force sensitivity can be enhanced by reducing the dimensions of conventional cantilevers by at least an order of magnitude. We have developed a mass-fabrication method for producing single crystalline cantilevers of typically 15x4x0.2 μm^3 (length x width x thickness), and resonance frequencies in the MHz range. To optimally benefit from these cantilevers, a Fabry-Perot interferometer has been built with a spot size of 3 □m on the cantilever at a working distance of 0.8 mm. The interferometer has been successfully tested on small cantilevers in vacuum, air, and liquid. As a next step, the small cantilevers and the interferometer will be mounted on a conventional scanner, to use them for high-resolution imaging of biomolecules.

Infrared nanocrystallography by near-field induced phonon-polariton resonance

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We exploit phonon-enhanced near-field interaction [1] in an infrared scattering-type scanning near-field optical microscope (s-SNOM) for mapping the structural properties of SiC crystals at nanoscale resolution.
Imaging of the SiC samples is done by a home-built s-SNOM in which the sharp probing tip of an atomic force microscope (AFM) is illuminated by infrared light from a tuneable CO2 laser (? ~ 9-11 □m). Along with the topography both amplitude and phase of the backscattered light are recorded thereby measuring the complex-valued optical near-field signal originating from the tip-sample near-field interaction. Close to the longitudinal optical phonon frequency (LO) of a polar material, the scattered light in s-SNOM exhibits a phonon-polariton resonance. The magnitude and spectral position of this resonance is extremely sensitive to the sample's local dielectric function ? which is correlated to the chemical and structural properties of the sample. By dipolar theory we predict that regions with equal chemical composition but differing crystal quality or lattice parameters can be distinguished by their infrared near-field spectra beeing an optical fingerprint of the material. This is experimentally confirmed by s-SNOM imaging of lattice damage in a 6H-SiC crystal induced by focused ion beam implantation (FIB)[2]. We also succeeded in differentiating 4H and 6H SiC polytypes at nanoscale resolution providing experimental evidence for the high sensitivity of our method.
Altogether, damping and shifting of the local phonon-resonance could be exploited for a quantitative, non-destructive and nanoscale-resolved investigation of crystal structures, useful for controlling implantation damage in semiconductor doping, lattice quality in crystal and thin-film growth, identification of crystal orientation in nanocomposites or quality control in wafer production.

[1] R.Hillenbrand, T.Taubner, F.Keilmann, Nature 418, 159-162 (2002).
[2] N.Ocelic, R.Hillenbrand, Nature Materials 3, 606-609 (2004)

SMS-suitability of new perylen and terrylen-based dyes and orientation of diffusing dyes in a molecular sieve by scanning confocal microscopy

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There is a longstanding interest in the synthesis and characterization of new fluorescent dyes. This interest is further increased by the possibility to use them as labels e.g. in biological systems. The duration with which single molecules can be observed is limited by properties like photobleaching and photoblinking. Most of the commercially available fluorescent dyes (e.g. Cy5, oxazine or rhodamine) have a poor photostability. On the other hand, more photostable dyes such as terrylendiimide (TDI) are of limited use in biological samples due to their poor solubility in aqueous media. New water-soluble perylen and terrylen-based dyes have been synthesized (Prof. Dr. K. Müllen, MPI Mainz). These dyes should combine an improved photostability, given by the perylen and terrylen structure of the chormophore, with an improved solubility in aqueous media, given by polar side chains. We developed a method to quantify accurately the photobleaching and photoblinking behavior. It allows to compare the performance of the new dyes with other ones (Oxazine-1, Sulforhodamine-B, TDI) in terms of their applicability towards SMS-based studies.

In a second study, terrylendiimide (TDI) dyes have been incorporated as guests into the pores of a spin-coated film of a mesoporous host-material (SBA-15). The orientational and translational diffusion of single TDI molecules were investigated simultaneously using confocal microscopy. The in-plane angle can be obtained with a precision of a few degrees. This method provides additional information for a more thorough understanding of the channel structure and its influence on the orientational and translational diffusion of the guest molecules.

Mediated Co-Adsorption and Stability of Grain-Boundaries of Hydrogen-Bound Molecular Monolayers at the Liquid-Solid Interface

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Self-assembled monolayers (SAMs) are an important grounding for future applications of long range ordered molecular structures in nanotechnology. Hence it is of general interest to understand the parameters determining the growth and stability of these systems.

By means of Scanning Tunneling Microscopy (STM) based experiments we try to learn more about driving forces for molecular self-assembly and the influence of external parameters.
Stable adsorption of TPT (1,3,5-tris(4-pyridyl)-2,4,6-triazin) molecules at the liquid-solid interface (which normally do not adsorb in an equilibrium situation) was observed by STM. Adsorption of TPT was made possible with the aid of H-bonding "glue-molecules" like TMA (1,3,5-benzene-tricarboxylic acid - trimesic acid) or TPA (1,4-benzene-dicarboxylic acid -terephthalic acid). With this method it was possible to prepare SAMs of TPT co-adsorbed with either TMA or TPA which were stable during the observation time of approximately 30 min.

Further investigations deal with the stability of H-bound molecular monolayers. Grain-boundaries of two different systems were observed for up to 20 min. The investigated systems exhibited great differences in their stability: two-dimensionally H-bound TMA monolayers were remarkably stable. Only a few fluctuations occurred in the vicinity of grain boundaries, but the grain size and shape was mainly preserved. The reason is that each TMA molecule is bound by a total of six H-bonds (two per carboxylic group), giving raise to the high stability. On the other hand grain boundaries of the one-dimensionally hydrogen-bound TPA system with it´s linear chains are quite unstable and one can observe rapid changes, like one domain growing on the expense of another one on a time scale of several minutes. The different behaviour is explained in the frame work of a molecular mechanics simulation, where an estimate of the binding energy of edge molecules was calculated.

Diffusion of Single Molecules in Nanostructured Molecular Sieves

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We investigate the translational diffusion of organic molecules in nanoporous materials using widefield-imaging microscopy and single particle tracking. The nanometre-sized channel system could be thought of a system of pipes in which molecules can diffuse. By analysing the trajectories of fluorescent dye molecules we can characterise the influence of the pore structure on molecular diffusion.
Two different studies are presented. First, terrylendiimide (TDI) molecules were incorporated into spin-coated mesoporous molecular sieves. Two pore architectures were synthesized: hexagonal (SBA-15) and cubic (SBA-16). It was possible to detect sub-populations of diffusing and non-diffusing molecules. In the hexagonal phase structured trajectories, reflecting the tortuosity of the channels, and unstructured ones of faster moving molecules could be distinguished. In contrast, the diffusional behaviour of TDI in the cubic material was found to be more homogenous. In the second study we compare the diffusion of a streptocyanine dye (9A1) in two types of sol-gel glass with average mesopore diameters of 3 nm and 20 nm. Significant differences in the diffusivity were observed depending on the diameter. Whereas in the smaller pores molecules showed changes between trapped and moving states, no such behaviour was found in the bigger pores.

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C. Hellriegel, J. Kirstein, C. Bräuchle, R. Brown et al., J. Phys Chem. B (2004), Published on Web.

Controlling spontaneous emission from colloidal II-VI nanocrystals in microcavities

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Colloidal II-VI nanocrystals are an interesting class of materials for optoelectronic devices. The solution-based synthesis of these nanoparticles typically results in a certain degree of polydispersity, which in turn gives rise to substantial spectral broadening of the ensemble. We demonstrate here that we are able to deposit composite nanocrystal/polymer films of excellent
optical quality, which can be used as the active layer in microcavities. These provide well-defined optical resonances, to which only a spectrally narrow ensemble of the nanocrystals couple, thereby resulting in an enhancement in spontaneous emission. We present data on time resolved fluorescence from these emitters embedded in microcavities and discuss the possibility of using the cavity induced field-enhancement to control energy transfer processes.

Immobilisation of recombinant proteins on solid surfaces - a method for single molecule biophysics

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A genetically modified form of the human DNA repair protein O6-alkylguanine-DNA alkyltransferase (hAGT) was used to immobilize various recombinant hAGT fusion proteins covalently and selective on gold and glass surfaces. The verification of the anchoring was done with SPR, AFM and fluorescence measurements. The results of these measurements prove the possibility to attach recombinant hAGT fusion proteins covalently, selective and directed on solid surfaces and do single molecule experiments on them.

High-resolution force spectroscopy with small Cantilevers for single molecular biological processes

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During the last years force spectroscopic measurements have contributed basic essentials to the clarification of pushing questions in the cell biology. This is due to the fact that the theory is in very good correspondence with experiments and one receives a quantitative insight into molecular interaction.
The purpose of the project is to improve the resolutions of an AFM for force spectroscopic measurements. This is possible by small cantilevers with higher resonance frequencies, which decreases the dominating thermal noise in the data. First measurements showed a 3 to 8 times better resolution, comparing to commercial cantilevers. Additional the resolution for the displacement is improved down to 1Å.
With a resolution in the sub-pN-area biological systems, like receptor-ligand systems and protein unfolding can be examined much closer and more exactly. Also measurements at biological transport systems, which possess at very low forces and small distances, are possibly.

Self-Assembly of Metal(II)-Phthalocyanines (Pc) and Naphthalocyanine (Nc)

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Ordered arrangements of organic molecules have received considerable attention in the context of sensors, optical devices, and molecular electronics. Scanning Tunneling Microscopy (STM) as a high resolution real space analysis is a unique tool to observe and even alter the arrangement of molecules on solid surfaces. In addition Scanning Tunneling Spectroscopy (STS) provides information about the local electronic density of states in the vicinity of the Fermi-level.

Here we compare the ordering of tin- and palladium-phthalocyanine for various coverages on different substrates. Both SnPc and PdPc form coexisting ordered and disordered phases on a Ag(111) single crystalline surface. In case of the non-planar SnPc different adsorption geometries of the shuttle-cock shaped molecule can be identified by means of high-resolution STM. The meta-stable ordered phase - found before tempering the sample - is comprised of alternating “up” and “down” oriented SnPc molecules. A statistic analysis of point defects was utilized to get an estimate for the difference in binding energy.

In order to reveal the influence of the size of the molecules on the monolayer unit cell the slightly larger Pc derivative naphthalocyanine was investigated on a graphite(0001) surface. The enlarged molecular structure mainly results in an up-scaling of a typical Pc unit cell. Moreover, reversing the tunneling voltage polarity showed a distinct difference in the submolecular STM contrast of occupied and unoccupied states. This difference is in good qualitative agreement with wavefunctions of frontier molecular orbitals as obtained by restricted Hartree-Fock calculations. Finally, STS gives information about the energies of molecular orbitals and is consistent with the experimentally observed difference in apparent height of occupied and unoccupied molecular orbitals.

Applications of Pulsed Interleaved Excitation in Fluorescence Cross-Correlation Spectroscopy

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Using pulsed interleaved excitation (PIE) and time-correlated single photon counting (TCSPC), we present a means of removing spectral crosstalk in fluorescence cross correlation spectroscopy. In our approach, we use a two-channel confocal microscope with two pulsed lasers (green and red) as excitation sources. The excitation pulses are delayed with respect to each other such that the detected fluorescence photons from one excitation pulse arrive before the following excitation pulse of the alternate color. Hence, the excitation source for each photon is known from the TCSPC signal. The cross-correlation function is calculated between photons detected in the green channel with green excitation and photons detected in the red channel with red excitation. Provided the green dye is not directly excited by the red laser and the appropriate spectral filters are used, the correlated signal will not suffer from cross talk and the minimally detectable cross-correlation signal improved. This technique can also provide accurate FCCS measurements in the presence of FRET. The method also promises to increase the accuracy of single pair FRET measurements by determining the stoichiometry of the fluorescent labels as well as correcting for changes in molecular brightness.

Microcontact Printing

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Patterns of Octadecyltrichlorosilane (OTS) or of the protein Streptavidin are printed onto a substrate using stamps of Polydimethyl Siloxane (PDMS). Thus cells can be forced to adhere only to destined regions. Biotinylated DNA-constructs can be attached to a stamped pattern of Streptavidin. The patterns are visualized using fluorescence microscopy.

Investigating the Conformation of Lactose Repressor using Fluorescence Resonance Energy Transfer

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The lac repressor is the regulating enzyme in the lactose digest of E.coli bacterium. It turns off the gene expression when lactose is absent through binding to two out of three operator sites within the lac operon. Thus, the enzymes responsible for the lactose digest, encoded by the lac operon, cannot be expressed.
The crystal structure of the lac repressor bound to an operator DNA is very well studied: The protein is a dimer of dimers, which forms a V-shaped structure in crystallographic studies, where each dimer has a DNA binding domain (1). The two dimers are linked by a flexible hinge and the crystal quaternary structure may not be the energetically most favorable structure in solution.
We investigated the distance between the two operator sites using single pair FRET. In our approach we used a 21bp DNA double strand containing the operator sequence Osym labeled with either donor or acceptor dye on the 5'-ends. The experiments were performed in the presence and absence of the inducer ONPF, which increases the stability of the lac repressor complex.
A general problem in single molecule FRET studies arises from a peak around zero efficiency caused by inactive acceptor dye (2). A combination of spFRET with pulsed interleaved excitation (PIE) allows determination of the FRET signal as well as the stochiometry.
We show the feasibility of using PIE in spFRET experiments. First results on the lac repressor system support the assumption of a more open form in solution.

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Giant Stark shift in single colloidal semiconductor nanocrystals

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Understanding the origin of blinking and spectral diffusion is a prerequisite for using single semiconductor nanocrystals as nanolabels, nano light sources or local ambient sensors. We show that spectral diffusion is strongly influenced by the shape of the nanocrystal. We have studied a novel class of nanocrystals consisting of an elongated CdS shell with an emitting spherical CdSe core located at one end of the shell [Nano Lett. 3, 1677 (2003)], which shows particularly stable single particle emission. The broken symmetry defines a spatial direction for surface charge movement along the shell towards and away from the core. The change in distance of the surface charges to the emitting CdSe core results in a continuous modulation of the local electric field strength and thus in a Stark shift of the emission energy. Additionally, a change of the linewidth of the single particle emission is seen, which is a signature of high frequency temporal dynamics of the surface charges. We observe a universal correlation between the linewidth and the emission energy during spectral diffusion. This correlation allows us to microscopically track the meandering charges on a nanometer scale from 5 K up to room temperature [J. Müller et al., Phys Rev Lett., in press].
Furthermore, application of external electric fields to single nanocrystals makes it possible to actively control the emission wavelength through the Stark shift.
The dependence of the fluorescence linewidth on the local electric fields provides a novel probe of the particle's nanoenvironment. Additionally, externally applied fields can shift the emission wavelength by more than 50 times the spectral linewidth at low temperature, which make these nanocrystals prominent candidates for single particle single photon optical modulators.

Synthesis and Characterization of Colloidal Nanocrystals

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In this work our goal is to study the properties of water-soluble nanocrystals. Since nanocrystals are synthesized in hydrophobic media, we used and developed three different methods to bring the nanocrystals into aqueous solution: the first method is called mercapto propionic acid coating and is based on the idea of ligand exchange. The second method is silanization. This method is based on the growth of a hydrophilic silica shell around the nanocrystals. And the third one is the polymer coating. Here the principle idea is basically to coat hydrophobic nanocrystals with amphiphilic polymers where the hydrophobic tails of the polymer intercalate with the tails of the hydrophobic coating of the nanocrystals and the hydrophilic part is pointing towards solution. But many biological reactions are not only based on aqueous solution but rather occur in electrolytic solution. Therefore, in order to use our nanocrystals for biological applications it is necessary that they are salt stable. For this reason we developed a method to determine the concentration of salt until which the Au nanocrystals are stable. We could show that for the polymer-coated particles this concentration was much higher than the one present in biological environment. Therefore our Au-nanocrystals can be used for any biological application. For CdSe and others nanocrystals another method must be developed. Furthermore the fluorescent properties of water solubilized CdSe and CdSe/ZnS particles were characterized. Although the particles remain fluorescent, the quantum yield is drastically reduced upon the transfer to aqueous solution. In the last part of this work, we studied how to bind our nanoparticles to other surfaces using biological molecules. For this purpose DNA molecules were used.

Probing lipid membranes and ion channels with high-frequency spectroscopy

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A coplanar waveguide defined on a glass chip is integrated within a microfluidic cartridge to allow for high-frequency testing of lipid bilayer membranes. The membranes are carefully deposited on the circuit using the vesicle fusion method. The quality of the lipid bilayer membranes is controlled by fluorescence microscopy. The deposition of the bilayers results in a significant change in the high frequency transmission properties of the circuit. We also embed alamethicin, a model ion channel forming peptide, in the bilayers in order to quantify the influence of channel charging events on the overall impedance. A combined setup for patch-clamp measurements in an on-a-chip geometry with high-frequency electronic access will be proposed.

Single Nanoparticle Sensors Improved by Gold Nanoshells

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The light scattering spectra of noble metal nanoparticles shows a distinct resonance due to a collective oscillation of the conduction electrons. The spectral position of this nanoparticle plasmon resonance inter alia depends on the particles' dielectric environment. The refractive index change induced by the specific binding of less than 200 streptavidin proteins to the surface of a single functionalized gold nanoparticle leads to a detectable red shift of the plasmon resonance [1].

Here we report how using single gold nanoshells instead of single solid gold nanospheres can improve single nanoparticle molecular sensors. Gold nanoshells consisting of a thin gold shell coated on top of a spherical, dielectric Au2S core are prepared following the synthesis reported by Zhou and Halas [2, 3]. We collect the scattering spectrum of an individual nanoshell illuminated in a darkfield configuration with a nitrogen cooled CCD camera attached to a grating spectrometer. Our measurements reveal a surprisingly narrow homogeneous NPPR line width of typically 180 meV for nanoshells with resonance energies of 1.8 eV. We deduce a surface scattering parameter of only 0.5 by comparing the measured homogeneous line widths and resonance positions of various single nanoshells with Mie-theory calculations. In further measurements, we show that Au2S/Au nanoshells respond much more sensitively than solid nanoparticles to changes of the dielectric constant.

In conclusion, gold nanoshells improve single gold nanoparticle molecular sensors threefold [4]: (i) their resonance position is in the tissue optical window where the absorption of tissue and whole blood is relatively low; (ii) they have narrow resonances which eases the detection of small NPPR shifts; and (iii) they show a higher sensitivity to changes in the dielectric environment.

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Towards molecular spintronics: Manipulating triplet formation and migration kinetics in conjugated polymers

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Electroluminescence in conjugated polymers has attracted broad interest for display applications. The efficiency of these materials is strongly affected by the spin dependence of the charge carrier recombination and large exchange interactions, leading to the formation of usually non-emissive triplet excitons. We recently developed a technique to visualize triplets directly by enabling strong, spatially localized spin orbit coupling, which permits room temperature radiative recombination in form of phosphorescence without modifying the triplet formation rate (Lupton et al., PRL 89, 167401 (2002)). Using this phosphorescence signature, we can image the diffusion of triplets directly (Reufer et al., CPL 381, 60 (2003)).
We are also able to address the process of triplet generation, which relies on spin-forbidden intersystem crossing from the singlet to the triplet state under optical excitation. As the conjugated polymer exhibits strong optical gain, the process of stimulated emission can be used to deplete the singlet state before intersystem crossing to the triplet state occurs. This allows us to image and time-resolve the process of triplet formation directly.
Strong external magnetic fields shift the balance between singlet and triplet emission under electrical excitation. With increasing magnetic field strength the phosphorescence yield increases. This effect may be attributable to a preferential spin orientation of the randomly injected charge carriers parallel to the magnetic field, thereby increasing the probability of triplet generation.

The Kinetics of a DNA-Based Machine that can Cyclically Bind and Release Thrombin

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A molecular machine that can be instructed to grab or release the human blood-clotting factor, α-thrombin, has been introduced recently. The machine is based on a known 15-base DNA sequence that adopts a chair-shaped conformation in the presence of potassium ions, which makes it bind strongly to α-thrombin. To this 15-base aptamer an extra sequence of 12 bases was added that controls the binding ability of the entire strand. Under normal conditions, this switch does not affect the α-thrombin-binding ability. But a second DNA sequence can trigger the switch by binding to it: when added to the mixture, it destroys the machine's conformation and releases α-thrombin.

The binding and unbinding of thrombin is observed by means of fluorescence anisotropy and fluorescence resonance energy transfer (FRET) measurements. Details of the α-thrombin release process are not yet fully understood. Kinetic rate constants can be extracted to a certain degree from FRET measurements at various concentrations and temperatures. To further investigate the kinetics of the machine we performed fluorescence correlation spectroscopy measurements with varied ratio of thrombin to DNA-machine in solution. Thus the dissociation constant of the binding of thrombin to the machine can be determined. A kinetic model for the release process of α-thrombin is developed and solved for certain assumptions.

Single Virus Tracing

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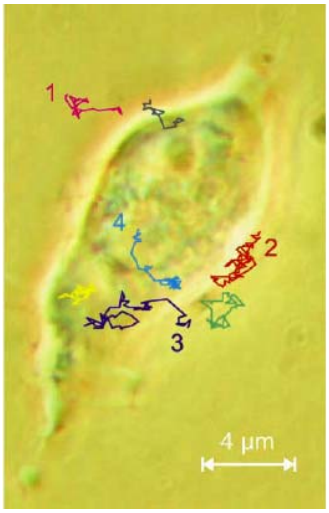
Viruses play a major role in biology and medicine. A detailed analysis of the different steps of a viral infection is not only necessary to understand viral biology, but also for the development of efficient antiviral drugs and secure vectors for viral gene therapy. Single Virus Tracing allows visualization of the infection pathway of an individual virus labelled with fluorescent dye molecules (even in the case of labelling with a single dye molecule). The fluorescence of the marker molecule is imaged and used to follow the pathway of the virus with high spatial (40 nm) and time (10 ms) resolution (Science, 294 (2001) 1929). As a first model system we have investigated the infection pathway of Adeno-associated viruses (AAV) into HeLa cells. The studies have been extended to other viral particles like murine CMV (Cytomegalovirus) and HIV.



The murine CMV is an enveloped virus, which was labelled at its core with GFP. First studies showed frequent membrane interactions, diffusion events in the cytoplasm and transport of a CMV-core on microtubules.

HIV is an enveloped virus, which was labelled at its matrix protein (MA) and its viral protein (Vpr) with GFP and GFP mutants. After analyzing trajectories of single HI-Viruses, we can give a detailed kinetic picture of the membrane interactions. Furthermore, using labelled fusion inhibitors, the inhibition of the membrane entry was observable.

Trajectories of single AAV-Cy5 particles indicating infectious entry pathways of AAVs into a living HeLa cell. (Science, 294 (2001) 1929).

DNA mediated dye/gold nanoparticle conjugates studied with time-resolved fluorescence

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Gold nanoparticles are efficient quenchers of fluorescence from dye molecules attached to their surface [1]. In the present study we have inserted carbon chains or single stranded DNA (ssDNA) as variable spacers between the dye molecule and the nanoparticle to investigate how the quenching efficiency depends on the distance between the two. The fluorescence lifetime rises markedly with growing spacer length. Varying the degree of ssDNA coverage we also observe that the fluorescence lifetime becomes significantly longer when the number of ssDNA per nanoparticle is increased. This gives evidence that the conformation of DNA bound to gold nanoparticles changes from wrapped to stretched when the surface coverage is increased [2]. The quantum yield of the fluorescent dye is reduced by 66% even for the greatest distance realized. This observation is in stark contrast to the enhanced fluorescence found near silver island films and silver colloid coated surfaces [3]. Together with our theoretical results for isolated particles it therefore suggests that fluorescence enhancement - similar to what is observed in SERS [4] - crucially depends on a field enhancement by several orders of magnitude, which is much higher near aggregated structures than near single nanoparticles.

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Combined single molecule force spectroscopy and fluorescence microscopy on organic light-emitting polymers

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AFM-based single molecule force spectroscopy is a useful tool to determine the elastic properties and the solvent behavior of polymers. Its combination with total internal reflection fluorescence microscopy (TIRF) provides a method to simultaneously visualize and manipulate luminescent polymers.

Through the AFM cantilever, mechanical force can be exerted on single polymer chains generating a force-distance curve. At the same time the luminescence of the polymer can be collected by using TIRF microscopy, allowing new insights into the physical properties of single polymers.

Depending on the solvent, polymers show characteristic behaviour. Polymers in good solvents are often stretched out, while in poor solvents they form random coils or aggregates. Both conformations react quite differently to stress which can be seen in differences in the force-distance plot.
The visualization of this behaviour requires polymers that are either labelled with a dye or emit light themselves.

Our research concentrates on organic light-emitting polymers e.g. the conjugated polymer Poly-[2-methoxy, 5-(2'-ethyl-hexyloxy)-p-phenylene-vinylene] (MEH-PPV) which is already known for its solvent-specific behavior.

Growth and structural characterization of pentacene on inert surfaces for applications in organic electronics

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Organic semiconductors have been the subject of intense research in the past decades because of their applications in electronic and optoelectronic devices. Although organic electronics are still far from, and actually not expected to be, replacing the high-end semiconductor devices, they are very promising in applications where elasticity, reduced cost, and ease of production are needed which cannot be provided by the current silicon based devices. Organic electronic devices generally employ thin films of the semiconductor material as the active region where charge is carried. Thus one needs to improve the film quality to get better device characteristics (e.g. large grain size, small number of charge traps).

To grow organic films, we use vapour phase deposition in a mobile uhv chamber. Base pressure of the deposition system, substrate temperature, deposition rate and the purity of the organic source material are the most important parameters, which determine the impurity concentration in the film and the adsorption dynamics of molecules on the substrate (e.g. diffusion length).

Among the various organic materials under investigation, pentacene (a long, flat, aromatic molecule) is particularly promising. It has been shown that pentacene forms layered crystals easily if deposited onto flat, inert surfaces, resulting in highly anisotropic transport properties.

To characterize the growth structure of these thin organic films, the combination of synchrotron x-ray diffraction and atomic force microscopy is well suited to study the early stage of film formation such as aggregation, molecular orientation, and layer-by-layer growth. Furthermore, one may study materials science aspects of these thin films such as grain size distribution and dislocation densities in detail.

To further improve the organic film quality, our current interest focuses on the growth behaviour of pentacene on top of organic templates such as silane or phosphonate based SAMs.

Coherent Infrared Spectrometer

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For improving the performance as well as spatial resolution of infrared microscopy, the conventional incoherent sources suffer from insuffcient focusable intensity. Coherent mid-infrared light can be generated from femtosecond laser pulses by means of difference frequency processes in nonlinear optical crystals, such providing a broadband illumination source for highresolution infrared microscopes. An application to the apertureless near-field microscopes developed in our group would allow imaging far beyond the Abbe limit.
The combined beam of two such sources with a slight difference in repetition rate constitutes a novel type of amplitude- and phase-sensitive Fouriertransform spectrometer dispensing with mechanical delay methods. This particularly allows a dramatic reduction of acquisition time to below 100 µs (1 µs anticipated) for a whole spectrum. Apart from microscopic imaging of spectral response, applications in remote sensing and single event analysis are possible with this spectrometer.

Investigating Protein Dynamics on Single Molecules by Total Internal Reflection Fluorescence Microscopy

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Since single molecules can easily be detected with optical fluorescence microscopy one of the emerging challenges in life and nanoscience is to investigate the dynamics of biomolecules on a single molecule level. Total Internal Reflection Fluorescence Microscopy (TIRFM) is an appropriate light-microscopic method to observe and analyze single molecule dynamics like protein motion or protein-protein interactions. The ultra sensitivity of TIRFM is due to the high s/n-ratio, which arises from illumination by an evanescent wave. Unlike confocal microscopy, TIRFM represents a method of widefield microscopy, so dynamic events of serveral single molecules at different sites can be observed simultaneously. By using TIRFM we are able to investigate single molecule dynamics of biomolecules which diffuses along DNA strands. We present a single molecule FRET-TIRFM experiment with a two color detection setup to analyze the molecular motion on the DNA strands.

Transport Measurement of Chemically Interconnected Carbon Nanotubes

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Carbon Nanotubes were opened to chemistry by introducing carboxylic groups to their ends and sidewalls, which allowed the attachment of biotin. The biotin-streptavidin reaction was then used to interconnect the functionalized nanotubes via streptavidin coated gold particles. These systems were lithographically contacted and transport measurement conducted.

Characterization of grafted polyelectrolytes with single molecule force spectroscopy

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Recent developments in the elaboration of inorganic/organic material have enabled the synthesis of novel polymer brushes possessing a wide range of tethered organic polymers onto silica surfaces (vinyl and acrylic monomers). Using controlled free radical polymerization conditions, linear chains were grown from the surface precluding an accurate control of the polymer monolayer thickness and polymer dimensions/structure.

AFM-based single molecule force spectroscopy employing an AFM allows for the determination of the surface and polymer characteristics of the interfacial architectures prepared by this 'grafting from'-approach. As a first system, grafted polymers with a pH-sensitive polyelectrolyte chain (polyacrylic acid, PAA) were studied because their Coulomb interactions can be effectively tuned by salinity and pH. Being able to visualize these tuning effects and assign e.g. lengths, single molecule force spectroscopy provides a useful tool for the investigation of polymers grafted to a surface.

Here, we present our results of desorption experiments of the interfacial polymer chains from silicon nitride tips as measured by AFM. The measured force curves reflect number, lengths and desorption forces of the substrate-bound polymer chains from an opposing silica surface. Most importantly, the lengths of the adsorbed polymer chains determined with force spectroscopy are in good agreement with previous molecular weight determination obtained by GPC. In addition, a pH-dependent shift of the experimentally acquired average polymer lengths indicates a conformational change within the surface-anchored polyelectrolyte chains.

Sub-microsecond molecular thermometry using thermal spin flips

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Thermal effects such as heat generation and dissipation are increasingly important in micro- and nanoelectronics and provide a substantial barrier to further miniaturisation. Molecular semiconductors such as conjugated polymers are often advocated as building blocks for nanoscale electronic devices, but the intrinsically low mobility of these materials implies that most of the electrical energy passed through the material is converted into heat. Consequently, there is considerable interest in techniques for thermal measurements on organic devices. We recently presented a highly sensitive molecular thermometer based on dual emission from the commonly used organic semiconductor platinum octaethyl porphyrin (PtOEP). This enables contact free optical thermometry of very thin films of conjugated polymers, such as in light-emitting diodes, and provides direct information on non-radiative decay channels in organic semiconductors (Appl. Phys. Lett. 81,2478(2002)).
We have now improved our detection scheme and are able to demonstrate the exceptional time resolution achievable with PtOEP molecular thermometers by measuring the instantaneous temperature of a conducting strip line on the nanosecond time scale. As the molecular thermometer works by thermally activated emission from a long-lived meta-stable state, we are able to achieve fluorescence based thermal imaging without the potentially perturbing influence of the exciting light source. Our method, which relies on gated fluorescence spectroscopy, therefore allows absolutely non-invasive characterisation of a totally thermally isolated system. Besides the technological implications and applications of adiabatic fluorescence thermometry to, for example, organic displays, the observation of externally triggered fluorescence bursts from organic semiconductors is of considerable fundamental interest. An external stimulus, in this case an electrically generated heat pulse, is used to flip the spin of the excited electron through reverse intersystem crossing from the triplet to the singlet state. Although the spins remain unpolarised, the control over spin correlation may provide a basis for molecular spintronic devices.

Nano-biological investigations on collagen for medical applications

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Collagen fibrils, type I and II with various periodicities were investigated, using the Atomic Force Microscope both as tool for imaging and nanomanipulation. Native and fibrous long spacing (FLS) collagen fibrils were formed by self assembling in vitro, using a special setup for dialysing. Depending on the conditions of assembly, collagen may form a variety of different structures (native fibrils, FLS fibrils, cocoon-like fibrils). The received collagen fibrils of type I had a bending pattern of 67nm for the native fibrils, and 200nm to 300nm for the FLS fibrils. Collagen is a system which shows a large degree of polymorphism. All fibrils with a periodicity greater than 67nm can be considered as FLS collagen. The collagen fibrils were imaged in air and in liquids. To determine the elastic properties of collagen fibrils the tip of the AFM was used as a nano-indentor. Force displacement curves were recorded in liquids at different heights of collagen clusters as well as on single fibrils and the Youngs modulus has been calculated utilizing the Hertzian theory. Results will be shown and discussed on single fibrils and on multilayer stacks.
To proove the feasability of this approach we carried out experiments on human heart valve sections. It was possible to determine different collagen assemblies in specific areas of the sections, that opens a wide field in pathological research. The discrimination of collagen assemblies in ventricular and atrial fibroelastica and fibrosa areas was possible. At the heart valve sections collagen type I and type II could be differentiated.
Furthermore this technique was used for the investigation of osteoplasts grown on different substrate coverings to show differences on compatibility of artificial implants to the growth of osteoplasts. An influence of the coverings to the growth could not be determined, but the homogeneity of the coverings were well defined. The BSP was more cristalline, wheras with fibrogen the surface was not covered homogenious, only collagen covered the substrate homogenious. The collagen acts here as an adhesive agent.

Miniaturization of biological assays by SAW driven planar fluidic networks

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The miniaturization of a lab down to chip size promises the advantages of low cost, high throughput and saving of resources. Employing surface acoustic waves (SAW) on the planar surface of a piezoelectric substrate, liquids can be stirred and moved along the surface. To realize a programmable “lab on a chip” it was demonstrated that the SAW driven actuation of small amounts of liquids yields a very attractive approach.
Mode conversion of SAW (Rayleigh mode) into bulk modes and vice versa opens the possibility to agitate and mix different liquids on separated from electric and electronic circuitry on the chip. For hybrid systems this setup can even be combined with nonpiezoelectric substrates like, e.g., silicon, glass or plastic.
For separation and analytical purposes, electrophoretic movement of molecules through a matrix is a common application in biochemistry. Amongst a variety of different electrophoretic techniques, the gel electrophoresis is probably best known . We demonstrate that such gel electrophoresis can be implemented into a SAW based “lab on a chip”. Combination of SAW based actuation and electrophoretic motion in a gel are combined to separate different ion solutions and more complex molecules on the chip.

Near-field infrared microscopy & nanospectroscopy of polymers

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Material discrimination of nanoscale composites can be a difficult task, because conventional optical and infrared microscopes are limited in resolution and other methods such as electron microscopy or AFM either lack contrast or are ambiguous. A new approach of near-field microscopy is capable of high spatial resolution and chemical sensitivity[1] given by an infrared spectrum.
We improved this scattering-type near-field infrared microscope (s-SNIM) to determine for the first time the near-field infrared spectrum of a 50-nm thin polymer film by tuning a CO-laser through a strong absorption band of PMMA around 5.8 um. The obtained near-field spectrum is characteristically different from the usual lorentzian shape of far-field absorption, but rather similar to a reflectivity spectrum. By imaging at chosen wavelengths to maximize contrast we identify PMMA in a 70 nm thin film of a nanoscale polymer blend. The smallest aggregates of 70 nm diameter are easily resolved, promising a resolution of less than 20 nm [2].

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Generation of Molecular probes for nanomedical applications

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Nanomedicine is the application of nanotechnology (the engineering of tiny machines) to the prevention and treatment of disease in human body. The discipline is in its infancy. It has the potential to change medical science dramatically in the 21st century. The most elementary nanomedical devices will be used for diagnosis. Chemical tests exist for this purpose; nanomachines could be employed to monitor the internal chemistry of the body. Here we present data on biomedical and biophysical methods to develop new nanotechnology diagnosis procedures. The combination of high resolution microscopy, such as atomic force microscopy (AFM), AFM nanomanipulaion and laser-based microdissection provides a direct approach for the investigation, isolation of biological specimen and the generation of specific molecular probes for diagnostic applications. Applications will be shown on different fields of medical and biological science: e.g. isolation of single chromosomes and chromosomal bands for cytogenetic diagnosis and gold-nanoparticle detection of region specific genetic samples. The possibility to miniaturize the diagnostic equipment in a lab on a chip system will also be discussed.

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Photoresponse of pentacene thin film transistors

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We investigate the photoresponse of pentacene thin film transistors. At applied source
drain and gate biases the photocurrent resulting from the illumination with white light of varying intensity and duration is measured, and the physical processes leading to this photocurrent will be discussed.

Spin correlations in polymeric semiconductors

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Conjugated polymers are a fascinating class of materials that combine the unique properties of plastics we know from our everyday lives with the electrical behaviour we normally associate with conventional inorganic semiconductors, such as their ability to emit light under application of a voltage. While organic light emitting diodes (OLEDs) have already entered the market in commercial applications, many fundamental questions concerning the nature of the excitations - strongly bound electron-hole pairs - are still unresolved. A detailed knowledge of these excitations is however important for optimizing device properties, such as the efficiency.
The use of a novel class of polymeric material containing traces of metallic complexes makes it possible to observe singlet and triplet excitons simultaneously for the first time, as the phosphorescent decay channel of usually invisible triplets is activated [1]. This way we can efficiently differentiate between spin states by spectroscopic means and directly observe changes of the total spin of the bound electron-hole pairs. We show that by applying an electric field, excitons can be stored as charge carrier pairs very effectively for time-spans exceeding their natural lifetime by several orders of magnitude. Using this technique we are able to provide experimental evidence for the previously postulated distant carrier pairs of triplet character for the first time. Astonishingly, during the electrostatic storage the total spin of the carrier pair does not change even at room temperature, suggesting an exceptionally strong exchange interaction. Such persistent spin conservation has not been expected in the literature up to now. In the contrary, by assuming fast spin-lattice-relaxation singlet-triplet ratios higher than 1:3 and therefore intrinsic OLED quantum efficiencies exceeding the statistical limit of 25% have been predicted [2]. Our results unambiguously show that this cannot be the case and that the efficiency of non-phosphorescent OLEDs must therefore be limited to 25%. Finally, we note that the strong exchange interaction at room temperature observed experimentally may open up new possibilities in the field of spintronics.

[1] J. M. Lupton et al., Phys. Rev. Lett. 89, 167401 (2002).
[2] M. Wohlgenannt et al., Nature 409, 494 (2001).

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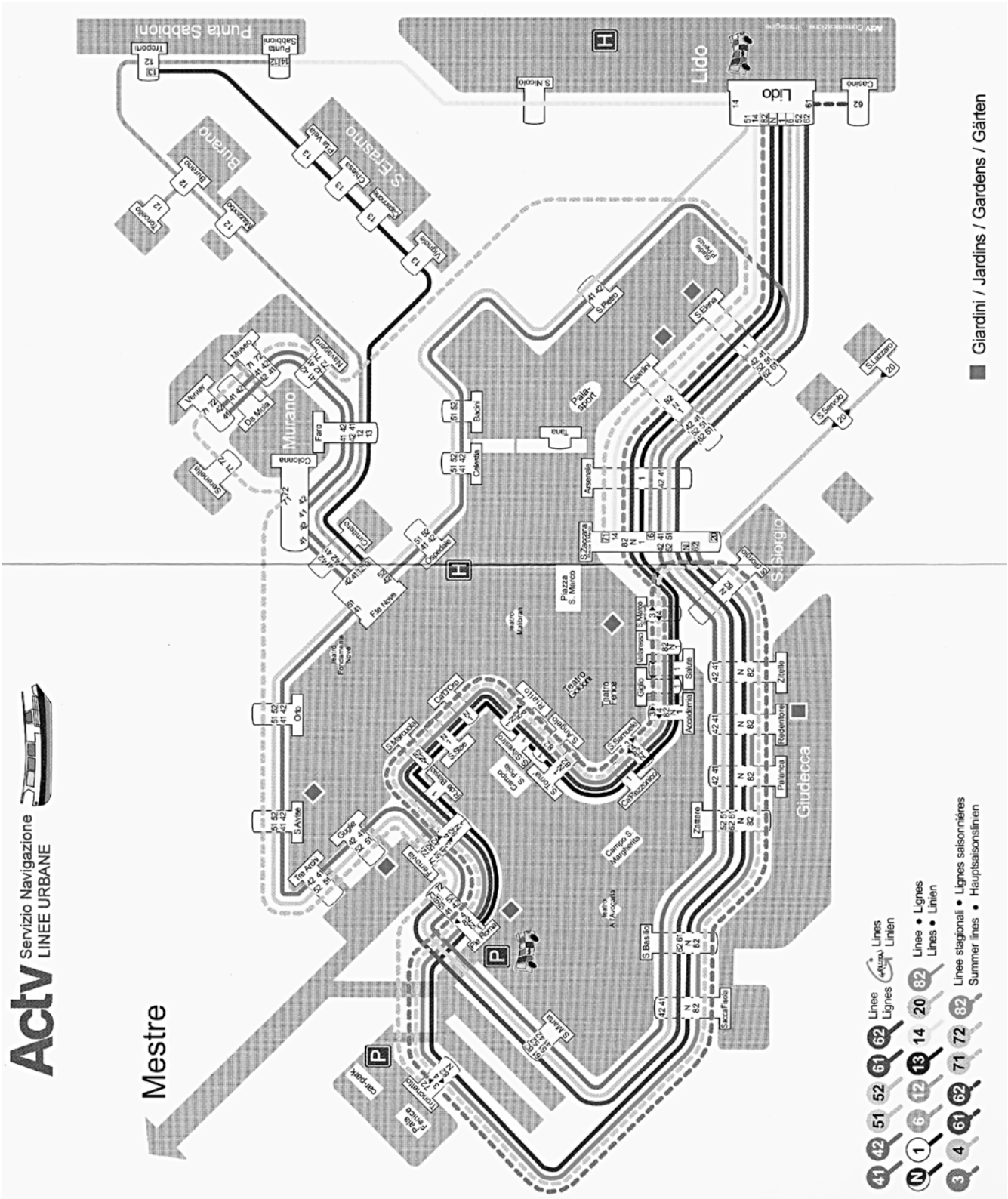

Transport in Venice

Boat Stop to San Servolo
BACINO DI SAN MARCO
CANALE DELLA GIUDECCA
CANAL GRANDE
CANAL GRANDE
CANAL GRANDE
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30124 Venezia
Tel.: 0039 041 5232333
Fax : 0039 041 5203721
fenice@fenicehotels.it, www.fenicehotels.it

The hotel is located very close to the Venetian Opera House.

2
Domus Ciliota
San Marco
Calle delle Muneghe 2976
in the city center, N.B. back by midnight
Tel. +39 041 520 4888
Fax +39 041 5212 730

Important: "Calle delle Muneghe" exists several times in Venice. Please make sure you head for the "Calle delle Muneghe" in St. Mark's quarter (Sestiere San Marco).
The Institute closes at midnight unless other arrangements are made.

3
Istituto Canossiano Pensionato Maria Immacolata
Dorsoduro 1323, 30123 Venezia
Fondamenta de le Romite/Eremite

Tel. +39 041 2409711/713
Fax: +39 041 2409712
E-mail: cvenezia@fdcc.org, giubileo@fdcc

The Institute closes at midnight unless other arrangements are made.
located close to the Accademia Bridge

4
Istituto "S.Maria della Pietà"
Calle della Pieta
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It is located very close to the boat landing for San Servolo.
The Institute closes at midnight unless other arrangements are made.

5
Hotel Palazzo Vitturi
Campo Santa Maria Formosa
Venice

From Piazzale Roma, take Waterbus n° 1 or 82, get off at Rialto stop, go along a few meters to Campo San Bartolomeo. Then Sottoportico della Bissa and walk along all of Salizzada San Lio at the end turn left, after you have gone over a bridge follow the canal side until you reach the Hotel.

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