

CeNS Workshop 2011

Nanosciences: From Molecular Systems to Functional Materials

September 19 - 23, 2011

Venice International University (VIU), San Servolo, Italy



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Artificial photosynthesis and heterogeneous organocatalysis by carbon nitrides

Markus Antonietti, Yong Wang, Xinchun Wang

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Nitrides and carbides are classes of nanomaterials which are presumably next on the scientific market to complement current functional oxide catalysts. Polymeric Graphitic Carbon Nitride (ideally C_3N_4) for instance is a graphite-like material, however with semiconductor properties and extreme chemical stability. Graphitic carbon nitride is however also chemically most surprising. Made from urea under early-Earth conditions, as shown already by Justus Liebig in 1832, it just recently turned out to be a novel catalyst which- among other reactions- can even chemically split CO_2 or photochemically turn water into hydrogen and oxygen. This opens the door to artificial photosynthesis

on the base of a sustainable and most abundant substrate base. I will introduce the basics of artificial photosynthesis on a tutorial base, go to required band structures and electronic schemes and couple that with chemical reactivity to catalyze both water oxidation and water reduction.

I will also present first schemes on chemical reactions where the electronic properties of C_3N_4 are generalized to other reactions, successfully mimicking oxidation enzymes with high conversions and selectivity.

Hybrid metal-semiconductor nanoparticles; From photocatalysis to doping

Uri Banin

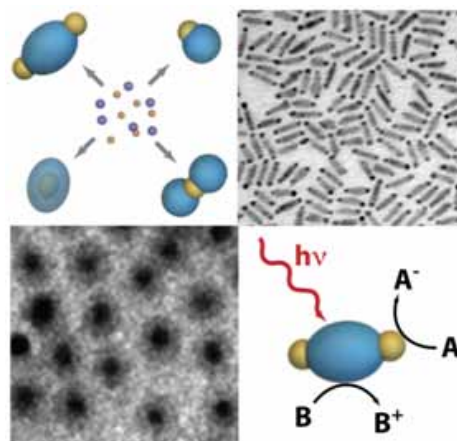
The Institute of Chemistry and the Center for Nanoscience and Nanotechnology, The Hebrew University of Jerusalem, Jerusalem, 91904, Israel

The ability to selectively arrange nano-sized domains of metallic, semiconducting and magnetic materials into a single "hybrid" nanoparticle offers an intriguing route to engineer nanomaterials with multiple functionalities or the enhanced properties of one domain. In this talk we will present recent strategies used to create semiconductor-metal hybrid nanoparticles, highlight the emergent properties of these multi-component materials, and discuss their potential applicability for different technologies.

One aspect of the multifunctionality of hybrid metal-semiconductor nanoparticles is related to light induced charge separation that was found to take place at the metal-semiconductor interface in such hybrids opening the path to their implementation in solar energy harvesting. Experiments examining the potential function of metal-semiconductor hybrid nanoparticles as novel photocatalysts will be reported. The formation of nano-cage hybrid nanoparticles as an interesting architecture for photocatalysis and sensing will also be highlighted.

Another aspect related to metal-semiconductor combinations is that of impurity doping in such colloidal nanocrystals which presents an important challenge. From the synthesis side, the introduction of a few impurity atoms into a nanocrystal which contains only a few hundred atoms may lead to their expulsion to the surface or compromise the crystal structure. From a physical viewpoint, impurities inherently create a heavily

doped nanocrystal under strong quantum confinement, and the electronic and optical properties in such circumstances are still unresolved. We developed a solution based method to dope semiconductor nanocrystals with metal impurities providing control of the band gap and Fermi energy. A combination of optical measurements, scanning tunnelling spectroscopy and theory revealed the emergence of a confined impurity band and band-tailing effects. Successful control of doping and its understanding provide *n*- and *p*-doped semiconductor nanocrystals which greatly enhance the potential application of such materials in solar cells, thin-film transistors, and optoelectronic devices prepared by facile bottom-up methods.



The first moments of STM and some thoughts about the future of nanotechnology

Gerd Binnig

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The very first moments of the development of STM and AFM with their major crucial decisions and results are described. Some conclusions are drawn from this historic consideration.

After a short detour into image analysis demonstrating challenges in biology and informatics some aspects of future potentials of nanotechnology seem to crystallize.

Chemical studies of genome maintenance and stem cell development

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A) Our genome is constantly damaged by various exogenous and endogenous events. 50'000 to 100'000 DNA lesions are generated each day per cell. The formed chemical DNA modifications interfere with normal DNA transcription and replication events causing mutations and cell death. In the lecture I will describe the chemical synthesis of DNA lesions that are formed by oxidative degradation processes. The DNA lesions are subsequently incorporated into oligonucleotides, which are crystallized together with DNA repair proteins and DNA polymerases. The results allow us to learn at atomic resolution of why these lesions are mutagenic.

B) In the second part of the lecture I will discuss of how the results can be used to create fully orthogonal new base pairs in which the hydrogen bonds that typically link the Watson-Crick base pairs can be replaced by coordination forces elicited by complexed metal ions and even reversible covalent bonds. The ability to use the tools of biology for the construction of artificial, self assembling covalent DNA structures has implications for the field of DNA based nano-systems.

C) Finally I will discuss that non-canonical nucleobases are also of key importance for developmental processes that lead to gene silencing and gene activation. During stem cell development

and neurogenesis, recently discovered new nucleobases such as hydroxymethyl-cytosine and formyl-cytosine play a pivotal role. I will discuss new synthetic routes to these compounds using modern metal organic chemistry and I will discuss how chemistry leads to new insights into the biology of stem cell development processes.

[1] G. W. Hsu, M. Ober, T. Carell, L. S. Beese, *Nature* 2004, 431, 217-221.

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[3] K. Tanaka, G. H. Clever, Y. Takezawa, Y. Yamada, C. Kaul, M. Shionoya, T. Carell *Nat. Nanotech.* 2006, 1, 190-194.

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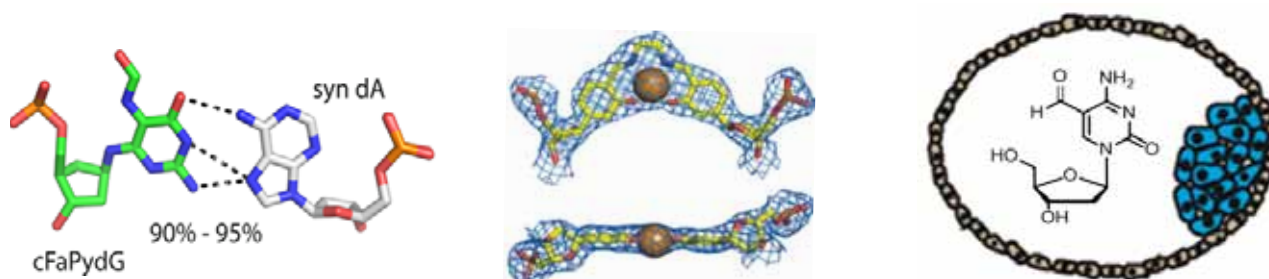


Figure 1: A: Mutagenic base pair of an oxidative DNA lesion. B: Artificial, covalently connected but still replicable metal base pair. C: The new base formyl-cytosine is a constituent of stem cell DNA.

Some like it hot – biomolecular interaction studies using microscale thermophoresis

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This presentation gives an overview on Microscale Thermophoresis, a new technology for biomolecular interaction studies. The term Microscale Thermophoresis refers to the directed movement of molecules in optically generated microscopic temperature gradients. This thermophoretic movement is determined by the entropy of the hydration shell around molecules. Almost all interactions and virtually any biochemical process comes along with a change in size, charge or conformation of a molecule that causes a change in the hydration shell. Such changes allow quantification of binding affinities of proteins, nucleic acids and small molecules. In addition also studies of complex molecular assemblies like

ribosomes and liposomes are possible. The flexible, fast and robust analysis of molecules directly in solution without surface coupling of molecules makes this method suitable for the analysis of molecules even in complex bioliquids like cell lysate. The method consumes only a few microliter of sample at nanomolar concentrations and measures affinities in the sub-nM to mM range. In this presentation we will describe technical details and benefits of Microscale Thermophoresis. We will show examples for interaction measurements ranging from protein-liposome, protein-protein, protein-nucleic acid, to small molecule-receptor interactions.

Hydrodynamics of microorganisms

Jörn Dunkel

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Hydrodynamics is essential for the self-propulsion of microorganisms, their mutual interactions and nutrient mixing in the surrounding fluid. Many structural properties of flagella or cilia (the microscopic appendages that propel bacteria and algae through their fluid environment) remain conserved in higher organisms, where they are known to play important roles in the reproductive system, during embryonal development and in the respiratory tract. Thus, studying the hydrodynamics of microorganisms not only helps to reveal the physical properties

of the simplest life forms, but also promises general insights that could be of medical importance in the future. In this talk, I will discuss recent experimental and theoretical progress in our understanding of the fluid flows generated by individual algae and bacteria. Specifically, I will present results from the first direct measurements of the *E. coli* flow field and discuss their implications for biofilm formation and collective behavior in bacterial suspensions.

Graphene quantum circuits

Klaus Ensslin

ETH Zürich, Laboratorium für Festkörperphysik, Zürich, Switzerland

Since its discovery in 2004, graphene has entered various different research fields. In the field of physics the special electronic properties of this two-dimensional honeycomb lattice have been studied. In contrast to conventional semiconductors, graphene has no band gap and charge carriers can be pictured as massless particles that obey the Dirac equation with the velocity of light replaced by the Fermi velocity. Additionally, the electron spin is expected to exhibit long coherence due to the weak interaction with the nuclear spins and with its own orbital motion. This property is promising for the realization of spin-based quantum bits as components of solid-state quantum computers.

Our work is mainly focused on the understanding of transport through nanostructures. Due to the absence of a band gap in single layer graphene, electrostatic confinement (as

implemented in e.g. GaAs devices) is not possible. It has however been shown that size confinement in etched nanoribbons introduces a transport gap. This feature has been utilized as tunneling barrier in graphene nanostructures such as single- and double-quantum dots. In recent experiments we have investigated the microscopic picture of transport mechanisms through these narrow constrictions in temperature dependent measurements.

In order to overcome the problem of the lacking band gap, bilayer graphene may be favorable. As the symmetry between the two graphene layers is broken, a band gap opens. This can be tuned by an electric field applied perpendicular to the layers. First experiments on double-gated bilayer nanostructures have been accomplished.

Voltage-controlled spin mechanics

Sebastian Gönnerwein

Walther-Meißner-Institut, Bayerische Akademie der Wissenschaften, Garching, Germany

Magnetic degrees of freedom are influenced by elastic deformations. In the "spin mechanics" scheme, one exploits this magneto-elastic coupling to realize an electric field control of magnetization.

On the one hand, we report on the voltage control of magnetization orientation in ferromagnetic nickel thin film/piezoelectric actuator hybrid structures [1]. The application of an electric field to the actuator results in a uniaxial elongation, which is directly transferred into the Ni film. Due to magneto-elastic coupling (inverse magnetostriction), this voltage-controlled strain modifies the magnetic anisotropy and thus induces a magnetization reorientation. In our spin mechanics hybrids, this allows for a voltage-controlled, fully reversible magnetization orientation manipulation within a range of approximately 90° at room temperature.

On the other hand, we show that the spin mechanics scheme also is operational at GHz frequencies. In the corresponding experiments, we use a surface acoustic wave (SAW) propagating in a Ni/LiNbO₃ hybrid device for the all-elastic excitation and detection of ferromagnetic resonance (FMR). Our SAW magneto-transmission data are consistently described by a modified Landau-Lifshitz-Gilbert approach [2], in which the magnetization precession is not driven by a conventional, external radio frequency magnetic field, but rather by a purely virtual, internal tickle field stemming from magneto-elastic interactions. This causes a distinct magnetic field orientation dependence of elastically driven FMR, which we observe in both simulations and experiment. Mechanically driven spin dynamics open interesting perspectives, e.g., for the study of FMR in magnetic nanostructures, or for spin pumping experiments.

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Cooperation, cheating, and collapse in microbial ecosystems

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Natural populations can suffer catastrophic collapse in response to small changes in environmental conditions, and recovery after such a collapse can be exceedingly difficult. We have used simple laboratory microbial ecosystems to study early warning signals of impending extinction. Yeast cooperatively breakdown the sugar sucrose, meaning that below a critical size the population is subject to sudden collapse. We have demonstrated experimentally that fluctuations of the population size can serve as an early warning signal that the

population is close to collapse. The cooperative nature of yeast growth on sucrose suggests that the population may be susceptible to cheater cells, which do not contribute to the public good and instead merely take advantage of the cooperative cells. We confirm this possibility experimentally and explore how such social parasitism can lead to population extinction.

Conformational dynamics in biological nanomachines: atomistic simulation and single molecule experiments

Helmut Grubmüller

Max Planck Institut for Biophysical Chemistry, 37077 Göttingen, Germany

Proteins are biological nanomachines. Virtually every function in the cell is carried out by proteins - ranging from protein synthesis, ATP synthesis, molecular binding and recognition, selective transport, sensor functions, mechanical stability, and many more.

The combined interdisciplinary efforts of the past years have revealed how many of these functions are effected on the molecular level. Computer simulations of the atomistic

dynamics play a pivotal role in this enterprise, as they offer both unparalleled temporal and spatial resolution. In this talk, using water channels as an example, the basic principles of the atomistic simulation are introduced. Subsequently, we'll have a closer look at the mechanico/chemical energy transfer in F1-ATP synthase, at the mechanical properties of viral capsids, and at close to atomistically resolved reverse translation steps in bacterial ribosomes.

Gap-mode plasmonic cavities: engineering light-matter interactions in metallic structures

Evelyn L. Hu, Kasey J. Russell, Kitty Yeung, Tsung-Li Liu, Shanying Cui

School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA

Optical cavities can tightly confine light in the vicinity of optical emitters, enhancing the interaction of light and matter. The modes or optical states of the cavity can be precisely designed and engineered, and in recent years there has been remarkable progress in demonstrations of cavity quantum electrodynamics (cQED) in solid state platforms. Such progress has been primarily for cavities fabricated in dielectric materials, with a steady improvement in cavity quality, with quality factors, Q , in excess of $10^4 - 10^6$ realized for cavities with coupled emitters. These high Q -coupled emitter systems have demonstrated heralded single photon emission, ultra-low threshold lasing and strong light-matter coupling.

Metal-based optical cavities would have inherently lower Q 's (and greater loss) than dielectrics; however, metal cavities utilizing surface plasmon polaritons (SPPs) can have sufficiently small mode volume to produce a substantial Q/V , the quantity

relevant for high Purcell factors, a measure of the light-matter interaction. This talk will focus on such *plasmonic cavities*, with optical modes formed within the gap of the two metal layers which defined the cavity. Initial structures are formed from silver (Ag) nanowires (NW), 70 nm in diameter and 1 - 3 microns in length, placed into close proximity to a Ag thin film substrate. Optically active material was interposed between the nanowire and the Ag substrate: we have studied PbS colloidal quantum dots, dye molecules and Alq3, a light emitting material that has been incorporated into organic light emitting diodes.

We will discuss the modes observed for these cavities, methods of tuning the cavity, and changes in the lifetimes of the emitters in the gap-mode cavities. The high Q/V possible for these cavities, and the range of organic and nanocrystalline emitters they can accommodate make these important building blocks for the exploration of light-matter interaction in the solid state.

Low-voltage organic transistors and circuits for flexible electronics

Hagen Klauk

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Organic thin-film transistors (TFTs) are of interest for applications requiring electronic functionality with low or medium complexity distributed over large areas on unconventional substrates, such as glass or plastics. Generally these are applications in which the use of silicon devices and circuits is technically or economically not feasible, such as flexible displays [1] and large-area sensor arrays [2]. Active-matrix displays based on high-efficiency organic light emitting diodes (OLEDs) [3] require TFTs that can be operated with voltages of about 3 V. A promising approach to organic TFTs that can be operated with such low voltages are gate dielectrics based on a thin, plasma-grown AlO_x layer in combination with an organic self-assembled monolayer (SAM); these hybrid gate dielectrics have a thickness of about 5 nm and a capacitance close to $1 \mu\text{F}/\text{cm}^2$ [4]. The static and dynamic performance of organic p channel TFTs with lateral dimensions around $10 \mu\text{m}$ is already sufficient for flexible OLED displays with VGA resolution, where the TFTs operate with frequencies of a few tens of kilohertz. However, to be useful for high-definition displays or high-performance integrated circuits, organic TFTs must be able to operate with frequencies well above 1 MHz, which requires scaling of the lateral TFT dimensions below $1 \mu\text{m}$. A promising technique for the fabrication of organic TFTs with such small dimensions at temperatures compatible with flexible plastic substrates is the use of high-resolution silicon stencil masks, which have led to the first demonstration of organic TFTs and organic integrated circuits operating with frequencies above 1 MHz at supply voltages below 5 V [5,6].

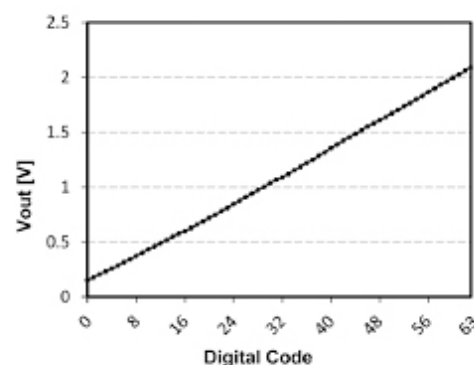
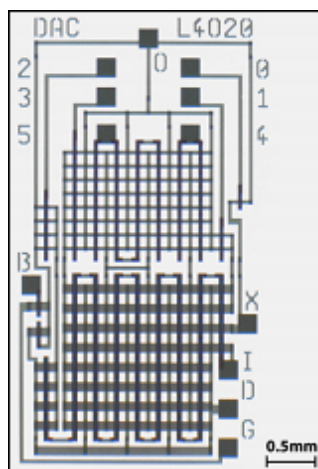


Figure 1: Photograph and transfer function of a 6-bit digital-to-analog converter (DAC) fabricated using low-voltage organic TFTs with lateral dimensions of $4 \mu\text{m}$. The DAC operates with a sampling rate of up to 100.000 samples per second.

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[3] G. He et al., *Appl. Phys. Lett.* 85, 3911 (2004)

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Solution based Carbon Nanotube films for electronics and optoelectronics applications

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Carbon nanotubes (CNTs) have been known for two decades and a lot of applications have been envisaged. Recently, it has been shown that CNTs can be treated like other organic materials and therefore be processed via solution-based techniques. The advantages of such technique are the low cost, the large area capabilities and the independence from the type of substrate. CNTs prepared in this way lead to a random interpenetrated conductive network that can have appealing applications in electronics and optoelectronics.

In this talk, we demonstrate a reliable and reproducible spray deposition process for the fabrication of CNT films exhibiting state-of-the-art performance. The convenient control of major process parameters enables a fine and accurate tuning of film characteristics, hence rendering this process suitable for a wide range of device applications with different requirements. Films, fabricated using this process, are applied, for instance, as resistive networks in gas sensing devices, as semitransparent electrodes in organic optical devices and as conductive layers of field effect transistors.

Due to the percolating nature of CNT films, the sheet resistance (R_{sh}) is expected to vary inversely with density or film thickness d . Figure 1a plots the relation between R_{sh} , as determined by four-point probe measurements, and the film thickness. One can clearly observe the reduction in R_{sh} with increasing film thickness, reaching as low as $60 \Omega/\text{sq}$ at approximately 42 nm. DC conductivity can be calculated in a straightforward manner from the sheet resistance and thickness through $\sigma_{dc} = 1/(R_{sh} \cdot d)$. The inset in Fig. 1a shows the calculated values of σ_{dc} as a function of d . As opposed to a material specific constant value for the conductivity, as expected from continuous metallic or semiconducting films, for a CNT thin-film shows a strong dependence on thickness. The conductivities calculated for different films vary by a factor of almost 4 in a range of thicknesses between 10 nm and 50 nm. As more CNTs are deposited, more conduction paths are created through the film and conductivity continues to increase until a thickness around 50 nm is reached, where film conductivity approaches saturation. In applications requiring optically transparent CNT films, the transmittance in the visible spectral range is of special interest. For this purpose, the transmittance at a wavelength of 550 nm is typically used as a

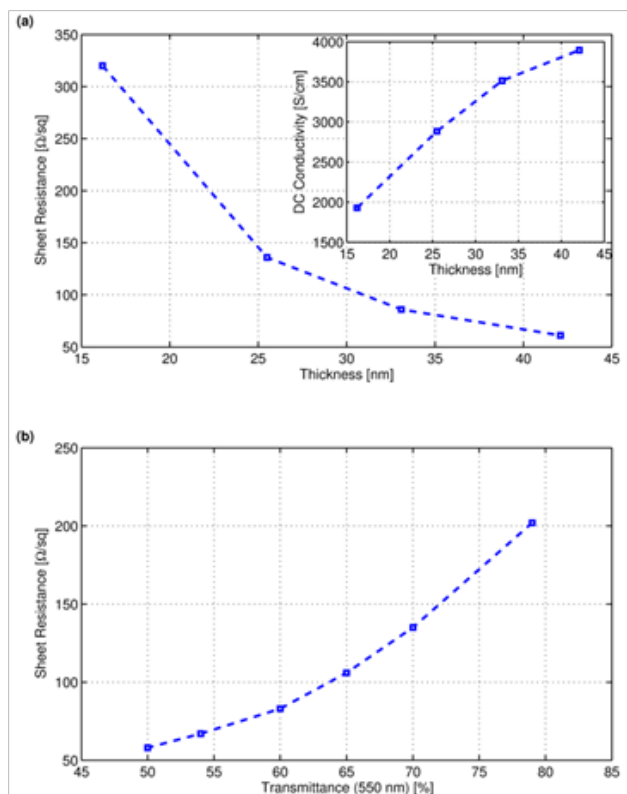


Figure 1: (a) Sheet resistance as a function of CNT film thickness; (b) Sheet resistance as a function of the transmittance at 550 nm.

representative value for comparison. A common approach for the evaluation of the electro-optical performance of such films is to plot sheet resistance as a function of the transmittance at 550 nm. This relation is plotted in Fig. 1b for the fabricated CNT films. A good trade-off between sheet resistance and transmittance is achieved with films having $200\Omega/\text{sq}$ at 80% or $105\Omega/\text{sq}$ at 65% transmittance.

The inherently high surface-to-volume ratio of CNTs renders them to be an ideal candidate for environmental gas sensing applications. A common implementation for CNT gas sensors consists of an interdigitated electrode structure forming the two contact terminals of the simple resistive architecture, above or beneath which a uniformly distributed resistive random network of carbon nanotubes is deposited. The sensor response is investigated by exposing the sensor module inside a gas chamber to different concentrations of ammonia (NH_3). The main figure used for the evaluation of sensor performance is the sensitivity, defined as the relative change in resistance during an exposure cycle. Figure 2a depicts the measured sensor resistance over time. The graph is divided into four segments, based on four exposure/recovery cycles at different NH_3 concentrations of 10, 25, 50, and 100 ppm. Each cycle is composed of an exposure interval followed by a recovery interval. One of the most prominent features observed in the response is the clear and immediate change in resistance at the start and end of NH_3 exposure. The magnitude of change in resistance after exposure to different concentration reveals a clear increase in sensitivity with increasing concentration, which becomes more evident when displaying the sensitivity as a function of NH_3 concentration, as plotted in Fig. 2b. Sensitivity is found to have a logarithmic dependence on the concentration, with sensitivities of 1% for concentrations as low as 10ppm and reaching 5% at a concentration of 100 ppm. We consider these values for sensitivity to be very competitive taking into account the short exposure intervals of 60 sec.

In addition, we will report on a similar process to fabri-

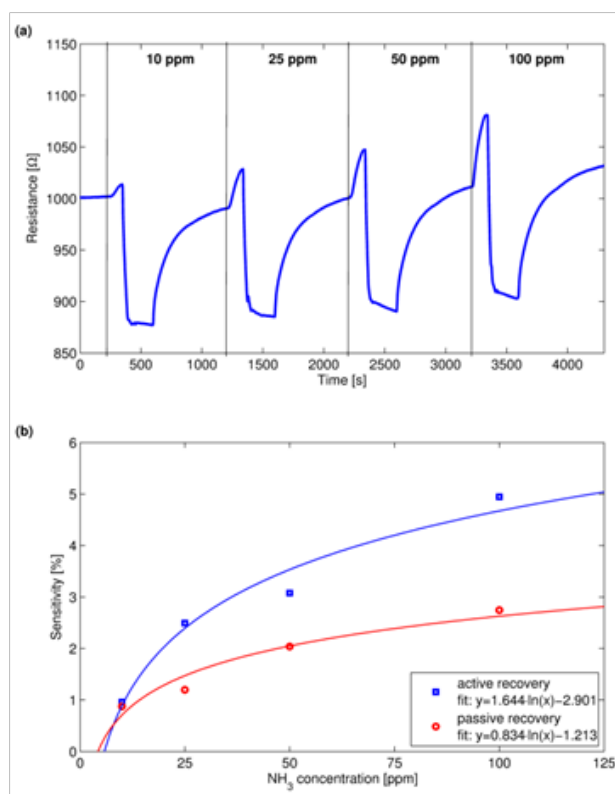


Figure 2: (a) Sensor response to NH_3 exposure; (b) Sensor sensitivity as a function of NH_3 concentration.

cate carbon nanotube field-effect transistors based on random nanotube networks. This solution-based approach is simple, fast and reliable, which can be applied to printed electronics. We investigated the transistor performance as a function of nanotube density, content of metallic nanotubes, and channel geometry. Our experiments show that increase of nanotube density results in consistent improvement of carrier mobility, until a threshold density is achieved.

A simple percolation model based on the Monte Carlo method has been developed for simulating the electrical characteristics of the CNT network. The model offers a basis for further optimization of carbon nanotube network devices. Each CNT is modeled as a "cylinder" with random length and direction. Junctions between CNT pairs are identified and determined by their types, which can be metallic-semiconducting, semiconducting-semiconducting or metallic-metallic (Fig. 3). Electrical segments are then defined as the nanotube portion that lies between two adjacent junction points, which form conducting paths between source and drain. After location and calculation of all valid "electrical segments", a HSPICE input circuit file is generated and executed, for calculating the total current of the CNT film.

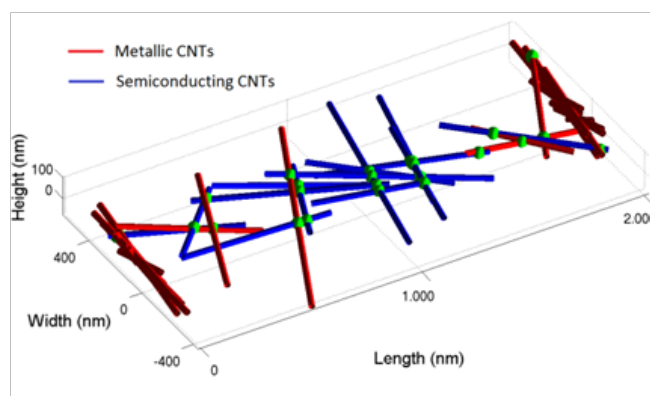


Figure 3: An example of random generated CNT network.

Order in graphene

Allan H. MacDonald

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Graphene is remarkable both for its unusual physical properties and for its elegant simplicity. Electrons in graphene are describable over a very wide range of energies by a two-dimensional Dirac like model in which the honeycomb lattice's sublattice degree of freedom acts like a pseudospin. I will also discuss three different unusual electronic states which may occur in graphene based two-dimensional electron systems, i) ferromagnets with an anomalous Hall effect and orbital magnetism but no spin-polarization, ii) excitonic superfluids and iii) states with a momentum space condensate that support dissipation-

less currents. All three states have unusual broken symmetries. Their properties are due to many-body physics that flows from the interesting variety of band-structures achievable when two or more graphene layers are coupled by inter-layer tunneling, or electron-electron interactions. There is now (still controversial) experimental evidence for the first of the three surprising broken symmetry states, while the second and third stand as unconfirmed theoretical proposals. I will review the status of this interesting new chapter in the strong correlation physics story.

Plasmon-assisted water oxidation on gold nanostructured TiO₂ photoelectrodesHiroaki Misawa¹, Yoshiaki Nishijima¹, Youzhan Zhang¹, Kosei Ueno^{1,2}, Kei Murakoshi³ and Haruo Inoue⁴¹ - Research Institute for Electronic Science, Hokkaido Univ., Sapporo, 001-0021, Japan² - PRESTO-JST, Kawaguchi, 332-0012, Japan³ - Graduate School of Science, Hokkaido Univ., Sapporo, 060-0810, Japan⁴ - Graduate School of Urban Environmental Sciences, Tokyo Metropolitan Univ., Hachiohji, 192-0397, Japan

Recently, we have demonstrated the plasmonic photoelectric conversion from visible to near-infrared wavelength by using electrodes in which gold nanorods (Au-NRs) are elaborately arrayed on the surface of TiO₂ single crystal. Significantly, there is a possibility that water molecules can serve as electron sources because stable photocurrents have been obtained for 200 hours, although only the electrolyte solution (KCl or KClO₄ aq.) was used in this photoelectrochemical measurement.¹ Here, we report on the study for elucidating possible electron sources in the photocurrent generation system to pursue the evolution of oxygen and hydrogen peroxide (H₂O₂) from the TiO₂ working electrode.

Au-NRs (240 x 110 x 40 nm³) showing localized surface plasmon (LSP) were fabricated on n-type TiO₂ single crystals (0.05 wt% niobium doped) with nanometric accuracy. Fig. 1(a) shows SEM images of the fabricated Au-NRs. This TiO₂ single crystal modified with Au-NRs was used as a working electrode. Photoelectrochemical measurements were performed using the three-electrode system. The evolution of oxygen and H₂O₂ was determined by GC-MS and absorptiometry using Oxo[5,10,15,20-tetra(4-pyridyl)porphinato]titanium(IV) as an indicator of H₂O₂, respectively.

Fig. 1(b) depicts the extinction spectra of Au-NRs prepared on the TiO₂ single crystal. A broad LSP band was observed around the wavelengths of 650 nm (transverse plasmon mode, T-mode) and 1000 nm (longitudinal plasmon mode, L-mode). From the action spectrum (Fig. 1(c)), the incident photon-to-photocurrent efficiency (IPCE) values were 6.2% and 8.4%, corresponding to the LSP bands in the T-mode at 650 nm and the L-mode at 1050 nm, respectively. It is noteworthy that the quantum efficiency of oxygen evolution upon irradiation from 650 to 750 nm is about 84%. This means that the chemical reaction which induces the oxygen evolution as a result of four electron oxidization of a water molecule proceeded almost stoichiometrically. Under the irradiation of light with a wavelength of 850 nm to 1150 nm, on the other hand, the efficiencies of the oxygen evolution and the H₂O₂ generation are 42% and 40%, respectively. Therefore, it is considered that two-electron oxidization of a water molecule has also occurred competitively with an irradiation of near-infrared light. Although the detailed reason why the efficiency of the H₂O₂ generation is different with the inci-

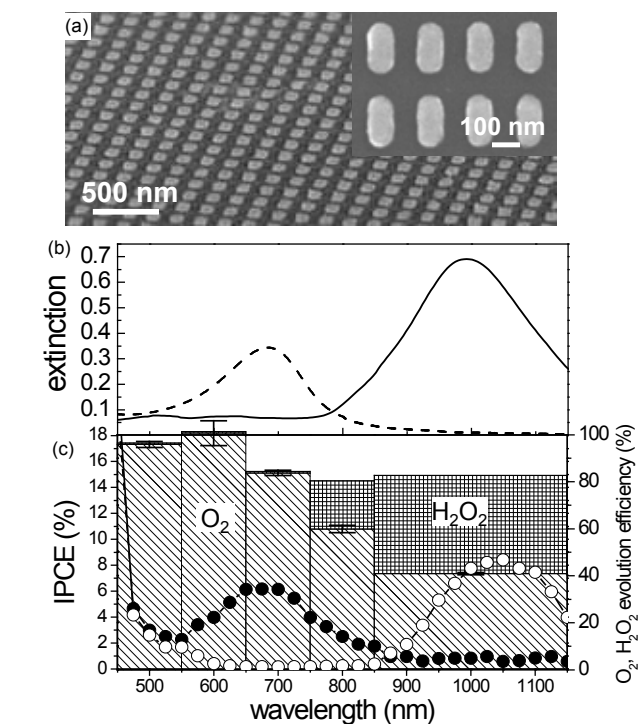


Figure 1: (a) SEM images of Au-NRs on TiO₂. (b) Polarized extinction spectra of Au-NRs; dashed line: T-mode and solid line: L-mode, respectively. (c) IPCE action spectra obtained by polarized light radiations; closed circle: T-mode, open circle: L-mode, and the relationship between the quantum efficiency of oxygen/H₂O₂ evolution and incident wavelength by an irradiation of broad band lights; 100 nm step from 450 nm to 850 nm, and 300 nm-wide wavelength from 850 nm to 1150 nm.

dent wavelength has not been understood in the current stage, it was verified that water molecules serve as electron sources in the photoelectric conversion system. The oxidation will be important because there is a possibility that the system can become an artificial photosynthesis system using near-infrared light radiation.

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Cell mechanics and the biophysics of membrane proteins

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Force nanoscopy can be used to undertake a multiscale approach to understand the mechanisms that underlie basic cellular processes, ranging from the cellular to molecular scale. I will introduce the use of atomic force microscopy (AFM)-based assays to characterize cell adhesion, cell migration, cell sorting and the dramatic shape changes of mitotic cells. AFM-based mechanical assays can be applied to measure the cellular and molecular interactions generated by these mechanical processes and to target individual membrane proteins playing com-

manding roles. Complementary AFM-based single-molecule techniques allow imaging membrane proteins at work and to probe the interactions and mechanisms that functionally regulate individual membrane proteins. Future developments of force nanoscopy, together with advances in light microscopy imaging and cell biological and genetic tools, should provide further insight into how membrane proteins contribute to basic cellular processes.

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An organic dual gate field effect transistor for biosensing

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Organic electronics aims on using soft matter materials such as polymers and small molecules for electronic devices. We explore under which conditions organic field effect transistors (OFETs) can be employed for biosensing. We show that concepts established originally for MOSFET technology can be transferred to organic electronics. In detail, we use a pentacene, the benchmark material for OFETs as semiconducting film. A thin film of long chain alkanes acts as top gate dielectric. An

analyte and a reference electrode act as top gate electrode. We show that the dual gate device geometry allows for tuning of the device into a region of high sensitivity. Micromolar sensitivity of such a device is demonstrated by the detection of stearic acid. Micromolar sensitivity is already sufficient for a range of medical applications such as glucose sensing in saliva. Thus, similar all plastic devices might be future alternatives for medical monitoring applications.

Non-equilibrium dynamics in graphene: perfect fluid behavior and stimulated emission of photo-excited fermions

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We present a theory for the non-equilibrium dynamics of monolayer graphene addressing the hydrodynamics near equilibrium and the ultrafast dynamics following an intense Laser pulse. The emergence of a quasi-conservation of the electron and hole numbers leads to a universal behavior of the d.c. conductivity and shear viscosity, making clean graphene an almost perfect fluid with tendency towards electron turbulence. The same quasi-conservation law leads to photoinduced femtosecond nonlinear saturation, transparency and stimulated infrared

emission of extremely dense fermions in graphene monolayers. We compare our theory for the transient state after an intense Laser pulse with recent experiments at high Laser intensity, explaining the observed perfect transparency in the nonlinear, high pulse regime. The underlying high-density yet stable population inversion at femtosecond times scales has significant implications in advancing graphene-based above-terahertz speed modulators, saturable absorbers, ultrabroadband gain medium and lasing tunable from the visible to terahertz.

Fundamental concepts and potential applications of strainable and shapeable nanomembranes

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In this talk conceptual opportunities and potential applications of inorganic and hybrid nanomembranes are presented. Nanomembranes become extremely flexible if they are not thicker than several tens of nanometers. They can be transferred from one substrate to another and shaped into almost arbitrary geometries. We produce nanomembranes out of semiconductors, metals, magnetic and organic materials and promote new functionalities and nanosystems both on and off the chip. This

includes stretchable magnetoelectronic devices [1], wavelength tuneable single photon sources [2], novel magnetic helical coils [3], lab-in-a-tube systems [4], hybrid semiconductor/organic heterojunctions [5], ultracompact energy storage devices [6] and multifunctional nanojet engines [7]. All these devices and innovations are possible by the mechanical elasticity of the materials and open a rich area of truly interdisciplinary research.

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Quantum simulation of frustrated classical magnetism in triangular optical lattices

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Magnetism plays a key role in modern technology and stimulates research in several branches of condensed matter physics. Although the theory of classical magnetism is well developed, the demonstration of a widely tunable experimental system has remained an elusive goal. We present the realization of a large-scale simulator for classical magnetism on a triangular lattice by exploiting the particular properties of a quantum system [1]. We use the motional degrees of freedom

of atoms trapped in an optical lattice to simulate a large variety of magnetic phases: ferromagnetic, antiferromagnetic, and even frustrated spin configurations. A rich phase diagram is revealed with different types of phase transitions. Our results provide a route to study highly debated phases like spin-liquids as well as the dynamics of quantum phase transitions.

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Self-assembled DNA-nanostructure tools for molecular biophysics

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Our group previously solved a key challenge for nanotechnology: programmable self-assembly of complex, three-dimensional nanostructures. Our solution was to build custom three-dimensional structures that can be conceived as stacks of nearly flat layers of DNA. I will discuss applications of this technology for molecular biophysics: (1) weak-alignment media for NMR structure determination of membrane proteins such as UCP2, a 30 kDa six-transmembrane helix mitochondrial proton

transporter; (2) scaffolds for determining the number of SNARE complexes required for lipid-bilayer fusion; (3) artificial actin filaments built as chimeras between protein and DNA for dissecting the mechanism of movement by processive myosin bipolar walkers.

Exciton dynamics in semicrystalline polymeric semiconductors

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Understanding charge generation by light absorption in polymeric semiconductors is of profound scientific importance due to the vigorous drive to develop organic solar cells. Confusion prevails with respect to the intrinsic charge photogeneration mechanism in neat (undoped) semicrystalline films. Numerous publications report charge photogeneration yields (the number of electron-hole pairs produced per absorbed photon) up to 30% on sub-picosecond timescales in neat regioregular poly(3-hexylthiophene) films. This is difficult to reconcile with the accepted picture that Frenkel excitons are the primary photoexcitations. Their binding energy is much higher than the lattice thermal energy at room temperature, such that direct charge generation ought to be improbable.

Considering this, two fundamental questions arise: (i) what is the mechanism of direct charge photogeneration in semicrystalline polymer semiconductors? (ii) What is the role of solid-state microstructure in defining it? Here, we combine transient photoluminescence and absorption probes and find that charge photogeneration at 10 K occurs continuously over sub-nanosecond timescales, and not by a diffusion-limited exciton dissociation at defect sites. Rather, we conclude that it is an extrinsic process that occurs efficiently by dissociation of excitons localised at interfaces between crystalline and non-crystalline domains, and is driven by interfacial energetic disorder.

Nanomechanical and microfluidic control over ion channels

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Ion channels are proteins that regulate numerous biological phenomena, from neuronal communication to cell osmoregulation to cardiac repolarization. Due to their wide-spread biological influence, malfunctioning channels are involved in a number of diseases. Currently, high-throughput drug-screening technologies are used to study ion channel biophysics under static conditions. However, ion channels *in situ* undergo dynamic forces, and replicating their natural environment requires precise, time-varying nanomechanical and microfluidic control. In this talk, I will outline technologies that show promise in realizing this more natural environment. One of these technologies utilizes a microstructured, piezoelectric quartz substrate for nanomechanical control over ion channels. These quartz substrates not

only yield lower noise than commercially-available technologies, but their piezoelectric properties allow for mechanical probing under either static or dynamic conditions. Similarly, microfluidic devices provide both static and dynamic control over the fluidic environment surrounding the ion channel. Of particular interest are high-speed microfluidic exchanges, which show promise in replicating natural synaptic communication on the nanoscale. Both microstructured quartz and microfluidic devices have been designed to easily integrate with current technologies. Therefore, high-throughput systems that study the dynamic responses of ion channels can now be realized.

Exploring the quantum physics of light with micro- and nanoelectronic circuits

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Using modern micro- and nano-fabrication techniques combined with superconducting materials we realize quantum electronic circuits to create, store, and manipulate individual microwave photons on a chip. The strong interaction of photons with superconducting quantum two-level systems allows us to probe the fundamental quantum properties of light. In particular, I will discuss experiments in which we realize an on-demand microwave frequency single photon source which we characterize by correlation function measurements. In the absence of efficient single photon counters, we use on-chip 50/50 beam splitters with off-chip linear amplifiers and quadrature amplitude detectors for which we have developed efficient methods to separate the detected single photon signal from the added noise [1]. We verify the operation of the single photon source by demonstrating single photon coherence and photon antibunching

in first and second-order correlation function measurements [2]. I will also present measurements in which we reconstruct the Wigner function of itinerant single photon Fock states and their superposition with the vacuum [3]. The techniques and methods demonstrated in this work may find broad application in the analysis of microwave radiation emitted from mesoscopic devices, in future linear optics and quantum information processing experiments.

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Structure and dynamics in actin networks

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In *in vivo*, the functionality of cells relies on a tight control of cytoskeletal actin structures which is obtained by a fine tuning of the actin binding proteins (ABPs). Each cellular process features its individual set of ABPs resulting in a well defined bundle width, optimized for this process. The underlying size limiting mechanism remains elusive. Using an *in vitro* model system, we show that size control relies on a mismatch between the helical structure of individual actin filaments and the geometric packing constraints within bundles. Furthermore, the dynamic reorganization of these cellular structures is an essential feature of cellular processes as e.g. cell migration. While various proteins are known to accelerate the polymer-

ization or disintegration of actin filaments, the insight in the mechanisms guaranteeing the kinetic stability of the cytoskeletal structures remains relatively scarce. We use multiple depolymerization methods to demonstrate that crosslinking and bundling proteins effectively suppress actin depolymerization in a concentration dependent manner. The presented results indicate that crosslinking ABPs do not only guarantee for the mechanical stability of a cell but also provide a powerful tool to stabilize distinct actin structures.

Life at the single molecule level

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In a living cell, gene expression - the transcription of DNA to messenger RNA followed by translation to protein - occurs stochastically, as a consequence of the low copy number of DNA and mRNA molecules involved. Can one monitor these processes in a living cell in real time? How do cells with identical genes exhibit different phenotypes? Recent advances in single-mole-

cule imaging in living cells allow these questions to be answered at the molecular level in a quantitative manner. It was found that low probability events of single molecules can have important biological consequences.

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Heparin-coated colloidal mesoporous silica nanoparticles as anticoagulant drug delivery system

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Colloidal mesoporous silica (CMS) nanoparticles attract great attention as potential injectable drug delivery system and in cancer cell targeting. In addition to their high biocompatibility and biodegradability, they can be selectively modified at their inner and outer surface.[1] Molecules can be efficiently encapsulated in their tunable pore system. Furthermore, they permit functionalization of their outer surface with targeting ligands, biomimetic and pore gating molecules, fluorescent dyes, and biocompatible polymers. Recent efforts have addressed their stabilization in aqueous buffers with poly(ethylene glycol) coatings.[2] However, to deliver such multiple core-shell functionalized CMS nanoparticles and to reach the targeted tissues or organs, additional efforts are critical in order to guarantee long circulation times and to prevent thrombogenic effects.

Here we present heparin-coated core-shell CMS nanoparticles (CMS-Hep), where heparin - a highly sulfated, anionic polysaccharide known for its anticoagulant properties - is covalently bound to the outer surface of amino-functionalized nanoparticles. This novel nanoscale system combines the efficiency of heparin in preventing blood-clotting with multi-functional core-shell CMS nanoparticles featuring high pore volume, tunable pore sizes and colloidal stability. Blood clotting tests and binding experiments with antithrombin (AT) proved the efficient anticoagulant functionality of the CMS-Hep. This novel nanocarrier offers new options for the design of drug delivery systems, for example in cancer therapy.

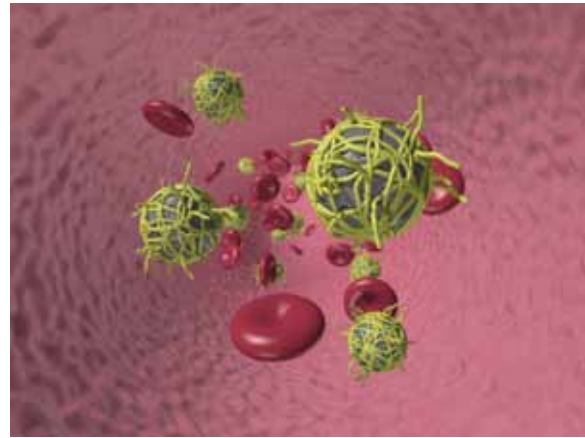


Figure 1: Schematic representation of CMS nanoparticles (grey) coated by an anticoagulant heparin shell (yellow) traveling in a blood vessel.

Acknowledgement

The authors are grateful for funding from the DFG through the SFB 749.

[1] V. Cauda, A. Schlossbauer, J. Kecht, A. Zuerner, T. Bein, *J. Am. Chem. Soc.* 2009, 131, 11361.

[2] V. Cauda, C. Argyo, T. Bein, *J. Mater. Chem.* 2010, 20, 8693.

Parasite motility: Mechanisms of a novel molecular motor, myosin XXI

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Myosin XXI is a motor found in the disease causing organism *Leishmania*. Genome analysis identified only two myosin genes, a class IB and a class XXI. While no expression of myosin IB has been found in the organism to date, myosin XXI has been detected in both the promastigote and the amastigote stages of the *Leishmania* life cycle, where it is preferentially localized to the proximal region of the flagellum.

The presence of only a single myosin isoform suggests that this myosin carries out a variety of functions within the protozoa, including possible roles in membrane anchorage as well as longer range directed movements with cargo. We aim to discover how myosin XXI can perform these different roles within the cell and how it is regulated to switch functions.

Using a baculovirus system we have purified a variety of constructs. We found that myosin XXI binds a single calmodulin at its neck domain and that this is required for motility, although not for ATPase activity. Myosin XXI transports actin filaments

in motility assays, and is insensitive to both high salt and Ca^{2+} concentrations up to $\text{pCa } 4$.

Sequence analysis of myosin XXI identifies a leucine zipper as well as two short coiled-coil regions, suggesting that myosin XXI is able to dimerise. To confirm this hypothesis we expressed a range of different length fluorescent tail constructs and studied formation and dynamics of dimerisation using gel filtration and a FRET assay. We found that the tail fragment (aa730-aa1051) binds calmodulin and dimerises in a concentration dependent manner. Intriguingly the full length myosin XXI initially appeared to be a monomer.

Further studies showed the tails dimerise in a temperature dependent manner. While the tails are stable monomers below $20\text{ }^{\circ}\text{C}$, they form dimers above this. When this was repeated with the full length protein we also observed the creation of dimers. We are now investigating if this dimerisation can be reversed in a physiologically relevant way.

Biophysical analysis of integrin specificity and crosstalk at the integrin-fibronectin adhesion junction

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Single cell atomic force microscopy (AFM) combines cell biology with the biophysical method to measure forces in the pico-newton range. This allows the analysis of forces between receptors on the surface of a living cell and its surrounding environment, e.g. the extracellular matrix (ECM). Cell adhesion and motility are tightly regulated by intra- and extracellular signals, which are communicated through the cell membrane by cell adhesion receptors. A major group of these receptors are force-transducing heterodimeric transmembrane proteins known as integrins. Although two distinct integrins, $\alpha v\beta 3$ and $\alpha 5\beta 1$, both bind the same extracellular matrix molecule, fibronectin, they evoke specific cellular functions. Still, the origin for those differences is assumably not only related to the integrins' cytoplasmatic affinity to distinct protein complexes, but also to their mechanical behaviour when conducting forces through the cellular membrane.

We analyze the mechanical environment of the two integrins, $\alpha v\beta 3$ and $\alpha 5\beta 1$, by measuring the unbinding forces of the integrin-fibronectin interaction with respect to the binding pocket and the intracellular anchorage. The model system used offers the unique opportunity to investigate either $\alpha v\beta 3$ or $\alpha 5\beta 1$ integrins on fibroblasts of the same genetic background. As exclusion of unspecific binding is a major challenge prior to access the fibronectin-integrin interaction, fibroblasts without a functional integrin are available to isolate the specific, integrin-related signals. Moreover, peptides and antibodies are available to specifically inhibit fibronectin-integrin interaction. We will present our data on integrin-specific adhesion patterns and unbinding forces for each of the two fibronectin-binding integrins, $\alpha v\beta 3$ or $\alpha 5\beta 1$, on fibroblasts.

Hydrodynamic deformation reveals two coupled timescales of red blood cell relaxation

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During their transport through the capillaries of blood vessels erythrocytes get repeatedly deformed many times. The elastic deformability of red cells in microcirculation is essential to maintain their shape and function for a life time of about 120 days. This enormous reversible shape deformability is based on the one hand on the non-spherical discoid biconcave resting shape and on the other hand on the mechanical properties of the lipid bilayer and the underlying cytoskeleton.

We have observed for the first time two modes of shape and rotational relaxation simultaneously in one single continuous

experiment by analyzing the relaxation of parachute shaped red cells. This allows us to consecutively measure both timescales corresponding to one individual red cell and to correlate the relaxation times. We find a linear relationship between the timescales and observe that with increasing deformation timescale also the timescale of rotational relaxation increase. We conclude that both timescales depend on the same parameter given by the mechanics of the cytoskeleton. Upon metabolic depletion and extracellular addition of ATP we modify the extent of cytoskeletal defects in the spectrin network and relate it to the mechanical properties of the red blood cell.

Optoelectronic properties of positioned InAs-based individual nanowires

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Walter Schottky Institut and Physik-Department, TUM Garching, Germany

Small bandgap semiconducting nanowires allow fabricating nanoscale light-sensitive devices like broadband solar cells or mid-infrared photodetectors. We discuss the optical and optoelectronic properties of positioned InAs based nanowires. To this end, p-Si(111) substrates with a top layer of SiO₂ are structured via e-beam lithography by holes with a diameter of approximately 80 nm. The nanowires are then grown vertically

on the substrates by solid-source molecular beam epitaxy. The optical properties of the nanowires are characterized by FTIR measurements. To fabricate optoelectronic devices, we subsequently embed the nanowires in an insulator (BCB). After an etch step, the nanowires are then contacted by depositing a thin conducting layer on top. The p-Si substrate provides the second electronic contact of the optoelectronic two-terminal devices.

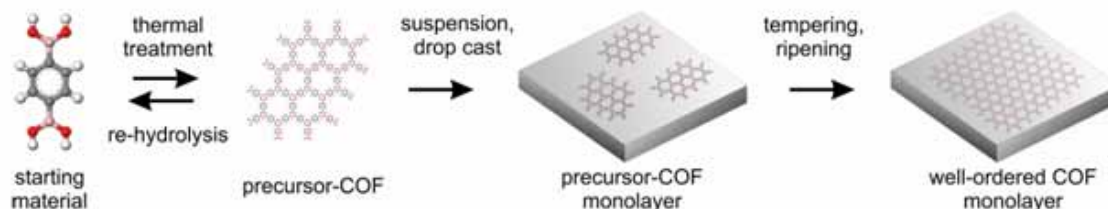
Synthesis of well-ordered COF monolayers via surface growth of nanocrystalline precursors

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Self-condensation of Benzene-1,4-diboronic acid can yield highly crystalline Covalent Organic Frameworks (COF). In the COF-1 structure, monomers are covalently interlinked into two-dimensional sheets which are then stacked along the c-axis. These sheets are comprised of a hexagonal alternating arrangement of phenyl and boroxine rings and are an ideal model system for 2D COF variants. In this contribution we demonstrate a novel approach to first synthesize nanocrystals of COF-1. The product of this synthesis was independently characterized by PXRD, TGA, IR-, and Raman-spectroscopy. Two-dimensional nanocrystalline flakes, whose structure corre-

spond to a single layer of COF-1, are then dispersed in a solvent and deposited onto an inert substrate. High resolution STM imaging under ambient conditions reveals full monolayer coverage of nanocrystalline COF-1 flakes with well ordered internal structure and lateral extensions of the flakes up to 10 nm. The lattice parameter of the COF-1 structures corresponds to the bulk value, and independently proves the synthesis of a covalent structure. Tempering of these highly polycrystalline structures in a humid atmosphere results in ripening, whereby the domain size of extremely well ordered covalent 2D structures increases up to 50 nm.

Determination of the electron diffusion length in dye sensitized solar cells by substrate contact patterning

Halina K. Dunn¹, Per-Oskar Westin², Daniel R. Staff³, Laurence M. Peter⁴, Alison B. Walker³, Gerrit Boschloo¹ and Anders Hagfeldt¹

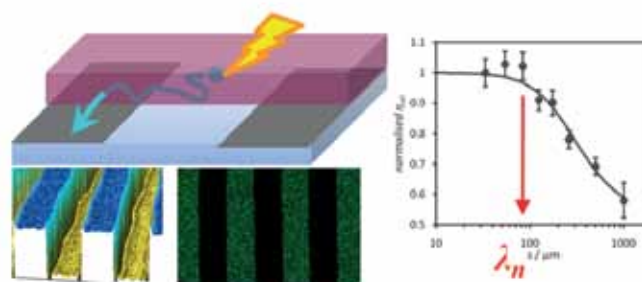
¹ Institute of Physical and Analytical Chemistry, Uppsala University, box 259, 751 05 Uppsala, Sweden

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A new method to estimate the electron diffusion length in dye-sensitized solar cells (DSCs) is presented. DSCs were fabricated on conducting glass substrates that were patterned by laser ablation of the FTO coating to form parallel contact strips separated by uncontacted strips of the same width. The relative collection efficiency was measured as a function of the gap between the contact strips, which determines the lateral distance traveled by electrons to reach the contacts. In order to avoid complications arising from non-linear recombination kinetics, current measurements were performed using small amplitude perturbations of the electron density close to open circuit and the maximum power point to minimize electron density gradients in the film. One and two dimensional solutions of the continuity equation for electron transport and back reaction predict that the relative collection efficiency should fall as spacing between the contact strips exceeds the electron diffusion length and electrons are lost by back electron transfer during transit to the contacts. Measurements of the relative collection efficiency were fitted to the predicted dependence of the collection



efficiency on the spacing between the contact strips to obtain the value of the electron diffusion length. The diffusion length is found to increase with voltage both at open circuit and at the maximum power point.

[1] Dunn HK et. al. *J. Phys. Chem. C*, 2011, 115 (28), pp 13932–13937

Electron amplification and phonon spectroscopy

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In state-of-the-art information technology, nonequilibrium electrons are essential. But they tend to relax their excess energy, and the emitted energy quanta can be reabsorbed by other circuit components. We therefore study the interactions of hot electrons in two-dimensional mesoscopic structures, considering a wide range of electron energies.

In our setup electrons are injected through an electrostatically defined barrier "BE" at a well-defined energy of eV above the Fermi energy, and then move ballistically along the sample. They transfer energy and forward momentum to electrons from the degenerate Fermi sea. A second barrier "BC" separates the collector "C" from the side contact. Excited electrons which carry enough forward momentum can pass the barrier and reach the collector contact, but positively charged holes (in the Fermi sea) are reflected [1]. If the side contact is grounded, the positive charge is neutralized by electrons flowing from the side into the device. This flow adds to the electron current from the emitter to create an amplified current at the collector port

Our analysis is based on an energy-dependent electron-electron scattering length as well as neutralization of holes.

Hot ballistic electrons not only scatter with electrons of the 2DES, but also emit acoustic phonons that can pass electrostatic barriers and be reabsorbed by electrons of the 2DES of an electrically separate circuit [2]. The resulting excited electrons have

a maximal kinetic energy, corresponding to maximum allowed energy that can be transferred by absorbing an acoustic phonon. This energy is measured using a third, calibrated, barrier "BD".

Measurements have also been performed for several charge carrier densities varied electrostatically via topgates as well as illumination.

[1] PRB 82, 161416 (2010)

[2] PRL 102,186801 (2009)

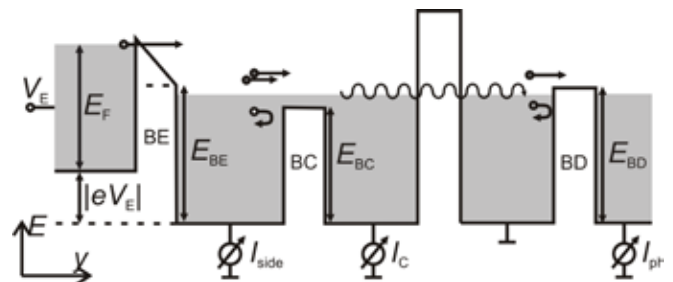


Figure 1: Energy diagram sketching the setup.

Red blood cell dynamics

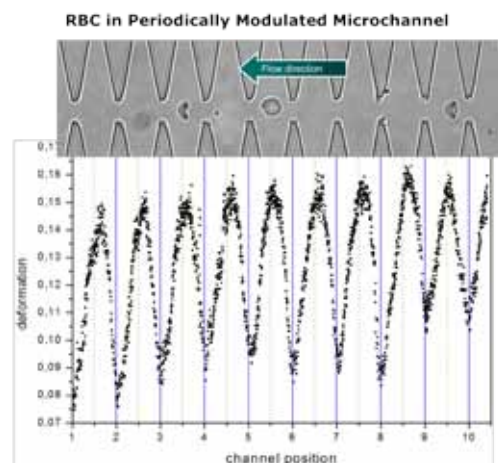
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The enormously reversible deformability of red blood cells (RBC) shape is based on both, the non-spherical discoid biconcave resting shape and the mechanical properties of the lipid bilayer with its underlying cytoskeleton.

Our microfluidic system studies systematically the elastic behavior of cells or vesicles that serve as a simple model system in hydrodynamic flow. Using a microfluidic PDMS channel with periodical width we are able to control frequency as well as amplitude of the external deformation excitation.

For RBC we find a transition from a regime with oscillating tilt angle of the main axis of the RBC and fixed shape to a regime with oscillating shape with increasing flow velocity. We have found the crossover to occur at a critical ratio of channel width and RBC velocity. These oscillations are superposed by shape transitions from a discocyte to a slipper shape at low velocities and a slipper to parachute transition at high flow velocities. For lipid vesicles we have observed three types of flow instabilities. At slow migration we found a transition from a state with orientational oscillations of a fixed prolate shape to a state with shape oscillations of symmetrical ellipsoidal or bullet-like shapes with increasing flow velocity as predicted by perturbation theory.



The transitions and their critical values are highly sensitive to the viscoelastic properties of the soft objects and therefore the method offers a simple and efficient approach to measure the cell mechanical properties of single as well as of large numbers of cells with excellent statistics.

Self-assembly of melem on Ag(111) – emergence of porous structures based on amino-heptazine hydrogen bonds

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Supramolecular self-assembly has been proven to be a straightforward and inexpensive bottom-up technique for surface functionalization. In particular, two-dimensional nanoporous networks that exhibit a regular arrangement of similar cavities have attracted broad interest. Size and shape of the cavities can be tuned by virtue of tailoring supramolecular building blocks. Many known porous structures are stabilized by intermolecular hydrogen bonds between carboxylic acids. Yet, on more reactive substrates, carboxylic acids often bind strongly to the surface, whereby formation of two-dimensional networks becomes hampered. Therefore, we investigate building blocks with less reactive functional groups for self-assembly of nanoporous networks on more reactive surfaces. In this re-

spect amino groups in combination with heterocycles offer a promising alternative. Here, we studied self-assembly of melem ($C_6N_7(NH_2)_3$), a triply amine functionalized heptazine ring, on Ag(111) by means of ultra-high vacuum Scanning Tunneling Microscopy. Seven different polymorphs were observed, where five structures exhibit similar pores with a diameter of ~ 1.5 nm, but variable interpore spacing.² Each of those pores is bordered and defined by six melem molecules which are interconnected by hydrogen bonds between amino groups and heterocyclic nitrogen atoms of the heptazine ring. While the structure of six out of the seven polymorphs can be described by a unified concept based on two intermolecular bonding motifs, a dense packed structure with different arrangement was also observed.

[1] B. Jürgens, E. Irran, J. Senker, P. Kroll, H. Müller, W. Schnick: Melem (2,5,8-Triamino-tri-s-triazine), an Important Intermediate during Condensation of Melamine Rings to Graphitic Carbon Nitride: Synthesis, Structure Determination by X-ray Powder Diffractometry, Solid-State NMR, and Theoretical Studies, *J. Am. Chem. Soc.*, 2003, 125, 10288–10300.

[2] J. Eichhorn, S. Schlögl, B. V. Lotsch, W. Schnick, W. M. Heckl, M. Lackinger: Self-Assembly of Melem on Ag(111) - emergence of porous structures based on amino-heptazine hydrogen bonds, *CrystEngComm*, 2011, Advance Article.

Picosecond time-resolved optoelectronic transport in nanoscale systems

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The time-resolved dynamics of photogenerated charge carriers in nanoscale systems are typically detected by optical techniques such as the transient absorption technique and the time-resolved photoluminescence spectroscopy. Many questions remain concerning the separation and the transport of photo-generated charge carriers to source and drain leads, when the nanosystems shall be functional modules in electronic circuits. Typical propagation times of ballistic photogenerated charge carriers in nanoscale circuits, which comprise semiconductor nanowires and quantum wires, are in the ps-regime [1,2]. Conventional electronic measurements cannot resolve such ultrafast dynamics because available electronic equipment cannot produce trigger signals and detect transients faster than tens of picoseconds. Furthermore, nanosystems typically exhibit a high impedance of several kilo-ohms, and ultrafast charge-carrier dynamics are therefore obscured by the response time of the high-frequency circuits. Here, we introduce an experimental on-chip scheme to mea-

sure the photocurrent dynamics of electrically contacted nanosystems in the time domain. The technique applies an ultrafast optical pump-probe scheme to a coplanar stripline circuit, and the photocurrent response of the nanosystems is sampled by a field probe (see Fig. 1 and [3]). The experimental setup with a picosecond time-resolution will be introduced, and first results of the ultrafast time-resolved photocurrent dynamics in contacted nanosystems, such as carbon nanotubes (see Fig. 2 and [4]) and graphene, will be shown. We will discuss polarization, plasmonic as well as charge transport effects within the nanosystems, the ultrafast relaxation of the photogenerated charge-carriers, and the influence of contacting the nanosystems by electrodes.

We gratefully acknowledge financial support from the German Science Foundation DFG (Ho 3324/4), FP7-Hysens, the Center for NanoScience (CeNS), the German excellence initiative via the "Nanosystems Initiative Munich (NIM)".

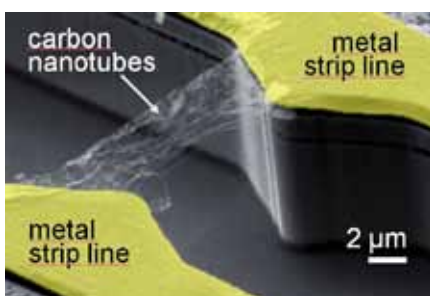


Figure 1: Scanning electron microscope (SEM) image of freely suspended single walled carbon nanotubes spanning two gold electrodes.

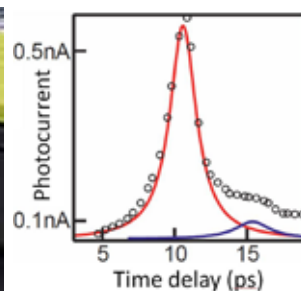


Figure 2: Picosecond time-resolved photocurrent of freely suspended single walled carbon nanotubes.

[1] C. Ruppert, S. Thunich, G. Abstreiter, A. Fontcuberta i Morral, A.W. Holleitner, and M. Betz, *Nano Letters* 10, 1799 (2010).

[2] K.-D. Hof, F.J. Kaiser, M. Stallhofer, D. Schuh, W. Wegscheider, P. Hänggi, S. Kohler, J. P. Kotthaus, and A.W. Holleitner, *Nano Letters* 10, 3836 (2010).

[3] L. Prechtel, S. Manus, D. Schuh, W. Wegscheider, and A.W. Holleitner, *Applied Physics Letters* 96, 261110 (2010).

[4] L. Prechtel, L. Song, S. Manus, D. Schuh, W. Wegscheider, and A.W. Holleitner, *Nano Letters* 11, 269-272 (2011).

3D-Nanostructured electrodes of transparent conducting oxides

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The development of nanostructured electrode layers with defined morphology is an important challenge in modern electrochemistry, as it enables the controlled design of complex electrochemical systems. Transparent conducting oxides (TCOs) play a special role in electrochemistry, being used as transparent electrodes for optoelectrochemical and electrochromic applications as well as spectroelectrochemistry. TCOs such as doped indium, tin or zinc oxides have been known for a long time in the form of dense flat layers obtained by physical deposition methods, but only recently the fabrication of 3D-conducting TCO networks has been reported. The interest in such 3D-electrode architectures is based on their large interface area enabling incorporation of large amounts of functional redox guests, with the electrical conductivity of the framework providing direct electronic access to the incorporated species, and their optical transparency allowing interactions with light.

We show fabrication of transparent conducting electrodes with various types and dimensions of 3D-nanostructures from different classes of TCOs, namely, antimony-doped tin oxide (ATO), niobium-doped titanium oxide (NTO) and indium tin oxide (ITO), by a directed self-assembly of corresponding nanoparticles [1-2]. Such transparent conducting matrices with defined porous architecture, high surface area and open accessible porosity can incorporate various redox moieties from small redox molecules to proteins and conducting bacteria films, which show the greatly enhanced electrochemical response proportional to the electrode surface area.

[1] V. Müller, M. Rasp, J. Rathousky, B. Schuetz, M. Niederberger, D. Fattakhova-Rohlfing. *Small* 2010, 6, 633.

[2] Y. Liu, J. Szeifert, J. Feckl, B. Mandlmeier, J. Rathousky, O. Hayden, D. Fattakhova-Rohlfing, T. Bein. *ACS Nano* 2010, 4, 5373.

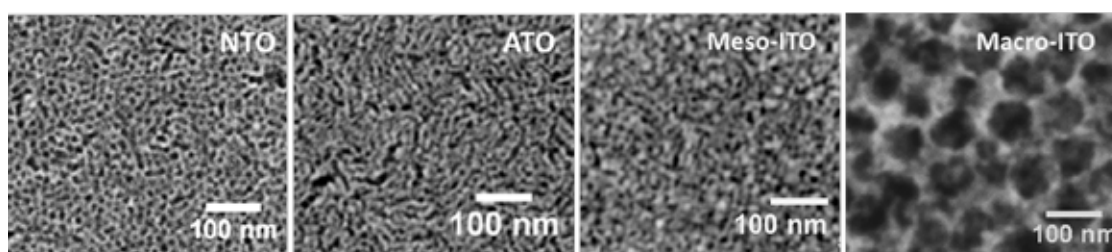


Figure 1: 3D-conducting transparent frameworks of different types of TCO materials and different dimensions of pore system.

Microwave cavity-enhanced transduction for plug and play nanomechanics at room temperature

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The readout and manipulation of nanomechanical systems can be significantly enhanced by coupling them to an optical or electrical cavity. The latter has the advantage of allowing to read out a large array of resonators by coupling to only one cavity without any positioning involved. Up to now experiments employing the coupling to microwave cavities are performed at cryogenic temperatures to benefit from superconducting cavities capacitively coupled to superconducting resonators and to minimize the dissipative effects of beam metallisation.

We present an approach based on a conventional lambda/4 microstrip cavity at room temperature with a resonance frequency of 3.5 GHz and a quality factor of 70. It is dielectrically coupled to a doubly-clamped high stress silicon nitride beam with an extremely high mechanical quality factor of 290,000 at a mechanical resonance frequency of 6.6 MHz. Displacement detection is performed by monitoring the mechanically induced sidebands in the microwave cavity transmission signal. The sensitivity is sufficient to resolve the Brownian motion. Furthermore, the obtained coupling is strong enough to observe backaction effects of the microwave photons on the mechanical resonator. We realize both cooling of the resonator fundamental mode to 150 K

as well as entering the regime of cavity-pumped self-oscillation. The latter generates a strong and narrow-band signal perfectly suited for sensing applications requiring a simple resonance frequency readout.

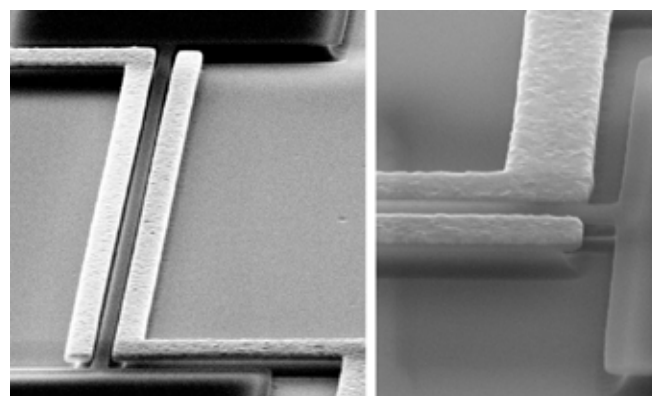


Figure 1: SEM Pictures of the silicon nitride beam (260 nm wide, 100 nm thick and 55 µm long) with the two adjacent gold electrodes

What encodes the diversity and ordering of self-organized monolayers?

C. Rohr,¹ M. Balbás Gamba,² K. Gruber,¹ E. C. Constable,³ E. Frey,² Thomas Franosch,⁴ and B. A. Hermann¹

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The a priori prediction of patterns formed from self-organized molecules is of crucial importance for the deliberate application of monolayers for surface functionalization. We used scanning tunneling microscopy (STM) to image self-organized monolayers of Fréchet dendrons, which display a large variety of 2D ordering motifs, even for a single molecule. We analyzed the ordering of the different phases of this high complexity system by molecular mechanics (MM) simulations. We conceived an elegant interaction-site model, which condenses the essential molecular properties determined by molecular mechanics modeling (MM), which in a Monte Carlo (MC) approach successfully predicts the various ordering motifs. With this model we could confirm that geometry as well as a few salient weak interaction sites fully encode the observed variety of structural motifs. Because of the simplicity of our approach it should be applicable to a wide variety of self-organized systems.

[1] C. Rohr, M. Balbás Gamba, K. Gruber, E.C. Constable, E. Frey, T. Franosch, and B.A. Hermann, *Nano Letters* 10(3), 833–837 (2010).

[2] B.A. Hermann, C. Rohr, M. Balbás Gamba, A. Malecki, M.S. Malarek, E. Frey, and T. Franosch, *Phys. Rev. B* 82, 165451 (2010).

[3] C. Rohr, M. Balbás Gamba, K. Gruber, C. Höhl, M.S. Malarek, L.J. Scherer, E.C. Constable, T. Franosch, and B.A. Hermann, *Chem. Comm.* 47, 1800 (2011).

Separation of blood cells via hydrodynamic lift

Thomas Geislinger, Benjamin Eggart, Susanne Braumüller, Lothar Schmid, Achim Wixforth, and Thomas Franke

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Our research is motivated by the attempt to develop Lab-on-a-Chip applications for medical analysis and sample preparation. Our experiments focus on the deformation and motion of soft objects such as lipid vesicles and red blood cells in hydrodynamic flow fields. The coupling of shear flow to deformation and motion of single soft objects gives rise to complex dynamic phenomena. Such dynamic phenomena can be the rotation of the fluid membrane around its interior ("tank-treading") or oscillatory ("swinging") and unsteady ("tumbling") motion. The observed regimes of motion and deformation critically depend on the membrane properties and the size of such soft objects. PDMS microchannels are used to compare the flow behaviour of red blood cells and blood platelets in simultaneous measurements. Our experiments demonstrate that red blood cells show a tank-treading motion and adopt a consistent height due to the hydrodynamic

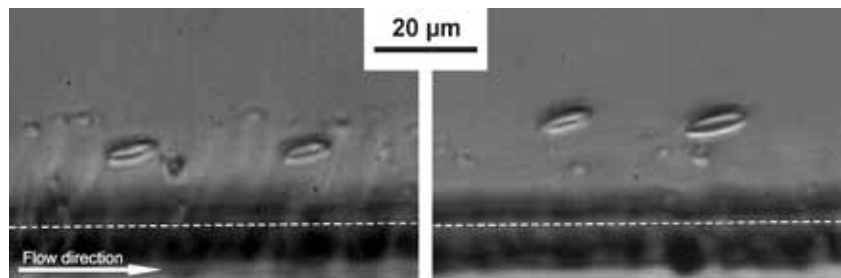


Figure 1: Red blood cells and blood platelets at the inlet (left) and at the end (right) of our microchannel. In the beginning, the cells all share a common height interval while they are separated from each other at the end of the channel due to the hydrodynamically induced lift.

lift-force and buoyancy. The smaller and more rigid platelets experience a weaker lift-force which leads to a lower adopted height at the end of our microchannels. Therefore, platelets and red blood cells can be separated from each other.

Permanent dipole moments of localized carbon nanotube excitons

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Semiconducting single-walled carbon nanotubes (CNT) show photoluminescence (PL) emission at near infrared wavelengths due to recombination of excitons [1-3]. Recent observation of photon anti-bunching in the PL of individual CNTs as well as asymmetric line-shapes of the PL emission spectra support the picture of exciton localization [4,5]. In a confocal microluminescence setup operated at cryogenic temperature we investigate the emission spectra of individual CoMoCat CNTs as a function of external electric field. We fabricated a sample that allows us to apply static electric fields perpendicular to the nanotube axis (Figure 1a). We observe spectral shifts up to several linewidths (Figure 1 inset) and a linear dependence of the emission wavelength on the electric field. We interpret our findings in terms of a linear DC Stark effect due to permanent

dipole moments of localized excitons. From the linear slope we deduce values for the exciton dipole moments as exemplified in Figure 1b for a specific nanotube with sub-Angstrom electron-hole-separation.

[1] M. J. O'Connell et al., *Science* 297, 593 (2002).

[2] J. Maultzsch et al., *Physical Review B* 72, 241402 (2005).

[3] F. Wang et al., *Science* 308, 838 (2005).

[4] A. Högele et al., *Physical Review Letters* 100, 217401 (2008).

[5] C. Galland et al., *Physical Review Letters* 101, 067402 (2008).

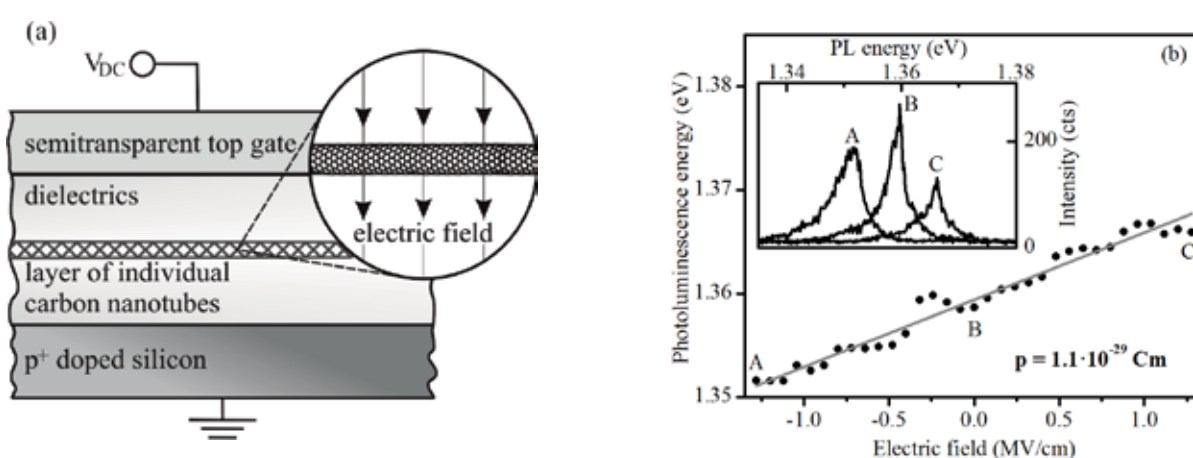


Figure 1: (a) Gate structure with carbon nanotubes sandwiched between dielectric layers and capacitor plates formed by a highly doped silicon substrate (back gate) and a semitransparent metal layer (top gate). By applying a DC voltage we reach electric fields E up to ± 1.2 MV/cm. (b) Photoluminescence energy shows a linear dependence on the electric field characteristic of linear DC Stark shift with a permanent dipole moment of $p = 1.1 \cdot 10^{-29}$ Cm. Inset: Photoluminescence emission spectra at -1.2 MV/cm (A), 0 MV/cm (B) and $+1.2$ MV/cm (C).

Investigation of TiO_2 layer in organic solar cell

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Organic solar cells (OSCs) based on poly-3-hexyl-thiophene (P3HT) and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) have attracted remarkable interest during the last decade, however, air stability of these cells remains poor. It can be significantly improved by using a thin electron-selective hole-blocking bottom layer like TiO_2 , which enables noble metals like Ag or Au used as top contacts, which are more stable.

In this study we investigate inverted TiO_2 layers fabricated by spray pyrolysis method as a blocking layer in OSCs, to investigate the influence of TiO_2 thickness on the power conversion efficiency and illumination stability of PSCs. Solar cells with a thicker layer of TiO_2 show lower short circuit current density (J_{sc}) and PCE, but higher stability. The dependence of performance on TiO_2 thickness is caused by a filter effect due to the trap states in our TiO_2 .

Forces across the scales: from cells to single molecules

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In implant research, there is a big necessity for a precise quantitative possibility to evaluate cell adhesion behaviour of different cell types on various surfaces consisting of different (altered) materials and geometric shapes. In the framework of the DFG-Project „Quantitative Evaluation der statischen und dynamischen Zelladhäsion und -aktivität an antibakteriellen DLC-Schichten“, we developed a surface acoustic wave (SAW) driven microfluidic system in order to investigate dynamic cell adhesion properties based on the knowledge of the shear-flow profile in the microfluidic chamber – forces on the macroscopic scale.

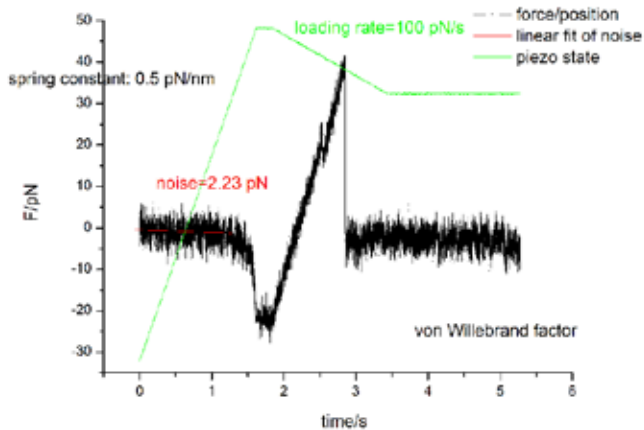


Figure 2: A typical single molecule stretching experiment (here with von Willebrand factor)

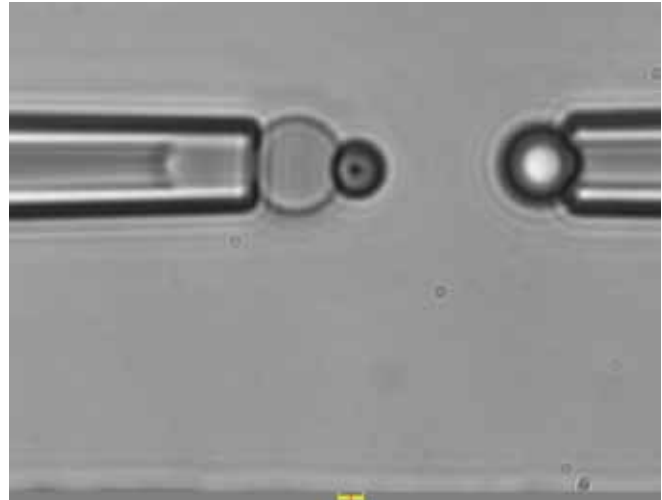


Figure 1: Top view into the BioForceProbe measuring chamber

On the microscopic scale in contrast, forces do not appear as a result of various effects, but are based on single molecule properties. For the investigation of them, a very sophisticated method for single molecule force spectroscopy is applied – the BioForceProbe (BFP) technique developed by Evans et al. 1995 [1]. Here, an erythrocyte in hypotonic conditions is used as a very soft spring. BFP allows to control both, spring constant and loading rates over several orders of magnitude. Further on, a very low noise level (~ 2.5 pN) can be reached.

[1] Sensitive Force Technique to Probe Molecular Adhesion and Structural Linkages at Biological Interfaces, E. Evans, K. Ritchie and R. Merkel, *Biophysical Journal*, vol. 68, 2580-2587, 1995

Klein paradox for arbitrary spatio-temporal scalar potential and Josephson-like current in graphene

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We derive the exact time evolution according to the Dirac-Weyl equation, describing a mono-layer of graphene, in the presence of a scalar potential $U(x,t)$ of arbitrary spatial and temporal dependence at normal incidence, $p_y=0$. This solution shows that the Klein paradox (the absence of backscattering) persists even for arbitrary temporal modulations of the barrier.

Moreover, we identify an unusual oscillating current j_y running along the barrier, despite of the vanishing momentum in y -direction. This current exhibits resemblance to the Josephson current in superconductors, including the occurrence of Shapiro steps and its sine-like dependence on the phase difference of wave functions.

Characterize and manipulate: tools to monitor bacterial interaction

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Letting bacterial strains of distinct traits compete against each other under pre-designed conditions allows real-time implementation of game theoretical approaches. Predicting the outcome of such a game and in a further step manipulating it, externally provoking victory, defeat or draw, requires knowledge of how to characterize key factors of bacterial interactions and of the effects that alterations of these factors entail.

In practice, we are looking at the Colicin E2 system of three fluorescence-labeled *Escherichia coli* strains representing a 'rock-paper-scissors' game: a poison-producing (C), a resistant (R) and a sensitive (S) strain provide a cycle with each strain having one antagonist being superior or inferior. The winner will always be the player eventually dominating the population by either killing or outgrowing the other contestants.

To be provided with a realtime indicator mirroring bacterial interaction, we investigated the correlation between fluorescence

intensity and growth rates of the bacterial players using flow cytometry and fluorescence microscopy for growth in fluid and on solid surfaces, respectively. In the course of this, fluorescence-activated cell sorting (FACS) proved to be a feasible method to analyze bacterial interaction. To specifically manipulate the 'rock-paper-scissors' game, we performed the above experiments also in presence and absence of different concentrations of Mitomycin C stimulating Colicin production, thus solely addressing the C strain. A directed alteration of bacterial interaction could be observed in solution as well as on solid surfaces.

The discovery of a decisive and quantitative correlation between bacterial growth rate and fluorescence intensity would provide a sensitive probing technique for the momentary bacterial fitness level resulting in a deeper understanding of bacterial interaction.

Synthesis and electron microscopy characterization of CuInS_2 films for photovoltaic application

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The development of new semiconductor materials for solar cells is a highly active area of research, aimed to simplify the manufacturing processes and to reduce production costs. In our work we want to design different nanocrystalline solar cells based on CuInS_2 as light absorber. This material is considered to be a promising candidate for solar cell applications because of its good long-term stability, its direct band gap of 1.53 eV which allows utilization of much of the solar spectrum and its high absorption coefficient [1]. By changing basic synthesis parameters, CuInS_2 can be obtained as an n- or p-type semiconductor [2].

In our research we focus on the synthesis of CuInS_2 thin films on fluorine doped tin oxide (FTO) substrates and their detailed microstructural investigation. Besides scanning electron microscopy (SEM) we use various transmission electron microscopy (TEM) methods. The latter include diffraction studies, conventional TEM, high-resolution TEM and high-angle annular dark-field (HAADF) imaging in the scanning TEM mode for crystal structure, morphology and grain size determination. The chemi-

cal composition analysis is achieved by energy-dispersive X-ray spectroscopy in the TEM and SEM.

We found that the processing parameters strongly modify the microstructure, ranging from spherical to more columnar structures. Thereby, thicker films showed sphere-like structures with sizes up to several μm and a stoichiometric composition. The CuInS_2 spheres consisted of individual flakes possessing grain sizes less than 100 nm. In contrast, thinner films showed two different regions. Close to the FTO substrate the film was densely packed while in the upper region columnar structures occurred, which were again formed by individual flakes. However, their grain size was much smaller and they were rich in copper compared to the ones forming the spheres.

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X-ray and neutron reflectivity study of tethered membranes

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Lipid bilayers chemically grafted to an interface provide a bio-functional surface to study lipid-protein systems. Here, in a multistep chemical reaction, PEG-2000 lipids were grafted to a silicon oxide surface via silanisation. Additional lipids, SOPC, spincoated to the surface form a bilayer when the sample is hydrated, which is tethered to the surface by the included PEG lipids. We have characterized the structure of the tethered lipid bilayers system with neutron and x-ray reflectivity. Using deuteration of selected components of the sample, to provide different contrasts in buffer and lipids, the hydration of the different

layers of the sample was determined. The PEG interlayer has a thickness of about 7 nm and is highly hydrated. At high pH, its thickness decreases by about 2 nm, which shows the flexibility of the PEG cushion. Diffusion measurements with FRAP showed a fluid membrane. The combination of the PEG as a soft and hydrated spacer and a diffusive lipid bilayer makes tethered lipid bilayers a suitable platform to include transmembrane proteins. They can be used to study the functions of a model cell membrane in a controlled environment of a flat surface in a fluidic chamber.

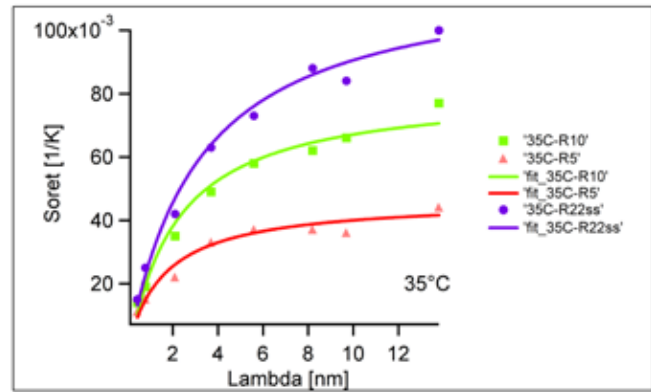
Curvature dependence in thermophoresis

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Thermophoresis, defined as the motion of particles induced by thermal gradients, has proven to be a valuable tool in analysing biomolecules. To test theories of thermophoresis, we measured the thermophoresis of highly diluted single and double stranded DNA and RNA using an all-optical capillary approach. The molecules are monodisperse and their characteristics are well known from biological studies. Temperature gradients were created locally by an infrared laser. The thermal depletion and back-diffusion of oligonucleotides between 5 and 50 bases in length were investigated by fluorescence end labeling at various salt concentrations. To a good approximation, the previously tested capacitor model describes thermophoresis. To show this we compared the concentration changes at different radii from the laserspot and over the a depletion and back-diffusion cycle with a numerical simulation based on the capacitor model. We investigated the dependence of thermophoresis on the temperature and arrived at the effective surface charge per base and characterized the hydration entropy resulting from the molecule - solvent interaction. Here we see an effective charge that decreases with the number of bases and shows similar

values for RNA and DNA. The Soret coefficient governing thermophoresis displays the transition from high to low curvature of the Debye layer, from spherical to planar condensator. Measurements of double stranded DNA and RNA show qualitatively comparable results.



Synthesis and characterization of biotemplated titania porous films

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Mesoporous titania films attract significant attention due to their successful implementation in photovoltaic devices, in particular dye-sensitized solar cells (DSC). One of the key issues in the performance of DSCs is the nanoporous morphology of the TiO_2 . Usually the organized porosity is achieved by using templating agents such as amphiphilic polymers. However, the tunability of the porous structures obtained in this way is limited. Nanocellulose (NC) consists of structural units of different dimensions lying in the mesoscopic range. The attractive features of NC are its shape anisotropy, its surface charge and an ability to self-organize. In addition, this novel material is obtained from abundant, "green" celluloses, which makes NC even more attractive in comparison with organic copolymers.

The current study investigates the influence of different amounts of NC on the morphology of obtained TiO_2 structures. The synthesis procedure starts by mixing NC suspension and the precursor solution (Figure 1). Subsequent film deposition promotes sol-gel and evaporation-induced self-assembly processes, leading to the formation of solid composite film. Finally, the composite is heated to remove the templating NC and to crystallize the titania matrix.

The biotemplated titania structures are characterized by sorption, Raman and X-ray diffraction measurements and electron microscopy. The obtained data show a very high potential of NC for introducing tuneable porosity into titania films.

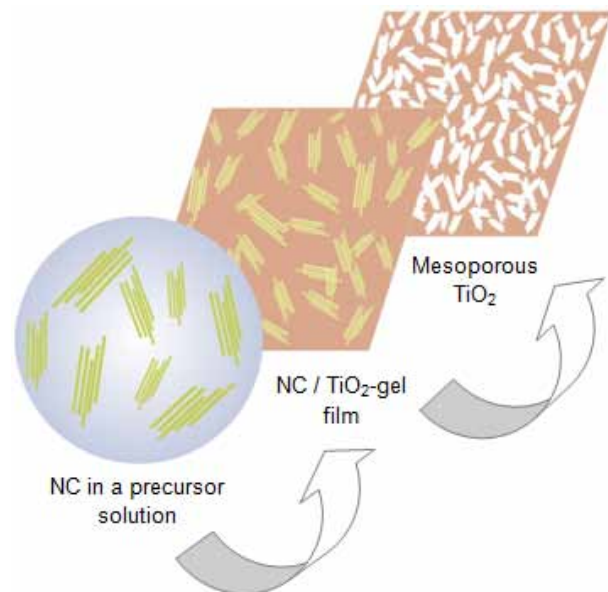


Figure 1: Synthesis approach to the biotemplated mesoporous TiO_2 films

2D and 3D image processing and analysis for the quantification of complex cellular features

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Bio-imaging unites the power of microscopy, biology, biophysics, biochemistry and advanced computational methods to study how genes, molecules and proteins work and interact in living cells and organisms. The newly-founded Bio-imaging Facility (established in 2009) provides access to high grade fluorescence research microscopes for scientists of the Gene Center to observe and analyze fixed samples as well as living organisms.

Image Processing

The use of well performing image analysis software is crucial to obtain quantitative and unbiased information. We provide of-line workstations for image analysis running various software platforms for 3D volume rendering and image analysis (e.g. Metamorph, Imaris and Definiens). Additionally, the Huygens software running on an 8 cores server can be used for deconvolution of the microscope images. This image processing technique improves dramatically contrast and resolution of the fluorescence images.

Magneto focusing of ballistic photocurrents in nanoscale circuits

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Quantum point contacts (QPCs) have recently been exploited in very sensitive detection schemes to quantify charge and spin states in nanoscale circuits and to image the coherent charge flow in two-dimensional electron gases (2DEGs) [1]. We demonstrate the use of GaAs-based QPCs to explore the non-equilibrium dynamics of photo-generated charge carriers in nanoscale circuits. We use QPCs to spatially resolve the ballistic flow of photo-generated electrons in a 2DEG [2]. To this end, electron-hole pairs are optically generated in a quantum well by interband laser excitation and the resulting optical beam induced current (OBIC) through an adjacent QPC is measured as a function of the laser spot position (Fig. 1). We observe that photo-generated electrons can ballistically propagate across several micrometers, before they tunnel through a QPC. Typical values for the exponential decay length of the OBIC amplitude along the electron paths are comparable to the electron mean free path $l_{mfp} = 15.1 \mu\text{m}$ of the 2DEG at 4.2 K.

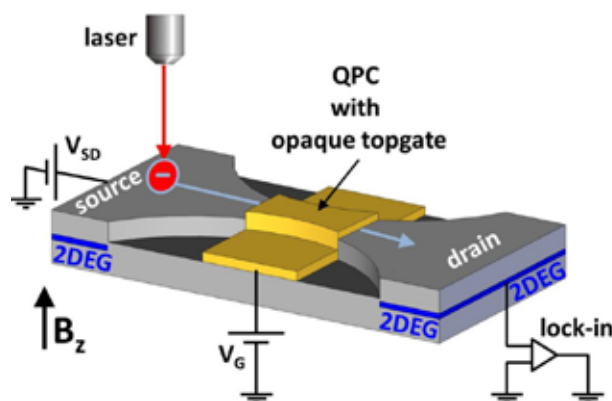


Figure 1: Optical beam induced current (OBIC) mapping in mesoscopic circuits: Schematic diagram showing a quantum point contact (QPC) with an opaque topgate (yellow) and optical excitation spot (red).

We further verify that the transmission of photo-generated electrons through a QPC is governed by the quantized energy and momentum values of the electron modes in the QPC. Hereby, the measured photocurrent across the QPC exhibits characteristic quantization steps as a function of both the topgate voltage and the source-drain bias [2].

In addition, we demonstrate that at a moderate magnetic field $|B_z| < 50 \text{ mT}$ applied perpendicular to the 2DEG plane, the photo-generated electrons move in cyclotron orbits. At higher magnetic fields, the electron motion is limited by the circuit geometry (Fig. 2).

We gratefully acknowledge financial support from the German Science Foundation DFG (Ho 3324/4), the Center for NanoScience (CeNS), and the German excellence initiative via the "Nanosystems Initiative Munich (NIM)".

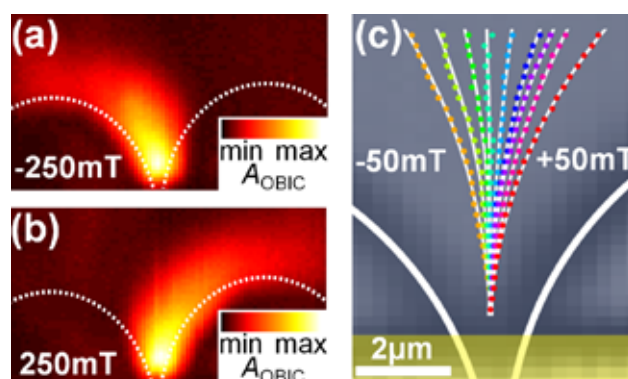


Figure 2: OBIC map of the source contact of a QPC at (a) $B_z = 250 \text{ mT}$, and (b) $B_z = -250 \text{ mT}$. (c) Map of reflected laser intensity from the source contact. Dotted lines show traces of the position of the maximum OBIC amplitude for different magnetic fields.

[1] M.A. Topinka, B.J. LeRoy, S.E. J. Shaw, E.J. Heller, R.M. Westervelt, K.D. Maranowski, and A.C. Gossard, *Science* 289, 2323 (2000).

[2] K.-D. Hof, F.J. Kaiser, M. Stallhofer, D. Schuh, W. Wegscheider, P. Hänggi, S. Kohler, J.P. Kotthaus, and A.W. Holleitner, *Nano Letters*, 10, 3836 (2010).

A low drift SPM with custom designed upgrades for self-assembly studies at liquid Solid Interfaces

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Studies of molecular dynamics at liquid-solid interfaces have the potential to contribute to our fundamental knowledge of supramolecular self-assembly. For this purpose, Scanning Probe Microscopes (SPM), like Scanning

Tunnelling Microscopes (STM) or Atomic Force Microscopes (AFM) are valuable and versatile tools. Here we present a low drift SPM with fully automated coarse approach, equipped with custom designed upgrades for studying dynamic and temperature dependent effects in monolayer self-assembly. This combined STM/AFM features easy exchange of probes, i.e. sharp metal tips for STM operation and a quartz tuning fork sensor for dynamic non-contact AFM mode.

Based on the open design of the microscope the standard setup can be extended by a heating stage facilitating temperature

dependent studies. Since entropic contributions are important in supramolecular self-assembly, temperature is an extremely important, but hardly addressed experimental parameter. The homebuilt heating unit allows to precisely control the sample temperature or to drive temperature programs by means of a feed-back controller. The design of the SPM allows for continuous imaging during heating in cooling cycles. This unique feature is demonstrated by temperature experiments at the liquid solid interface, where thermally induced desorption and re-adsorption of monolayers are observed in real space with molecular resolution.

We also demonstrate that the SPM design is suitable to study dynamic processes in supramolecular host-guest systems. By means of an injection system, additional solution can be injected under visual camera control while scanning without impairing the STM resolution nor imposing lateral drift. This experimental extension was successfully applied to study the incorporation dynamics of molecular guests into supramolecular host systems. In these novel experiments, we could show that not only the geometric match between guest and host pore, but also the solvent plays a decisive role.

Role of XPA in nucleotide excision repair

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Nucleotide excision repair (NER) is the principal genome maintenance system in mammals. The repair system presents a promiscuous lesion recognition mechanism that enables the removal of DNA lesions caused by UV-radiation or other environmental carcinogens. A defective NER response in mammals leads to a high predisposition to skin cancer (*Xeroderma Pigmentosum*). By incision of the damaged strand on both sides of the lesion the damaged oligonucleotide can be removed. XPC is known to detect disrupted duplex structures whereas XPA

is thought to be required for the recognition of bulky adducts generated by reaction of metabolically activated aromatic compounds. Since the exact mechanism of the DNA damage recognition is not fully understood, we first investigated the binding of the human protein XPA and its yeast homolog Rad14 to DNA lesions with EMSA (Electrophoretic Mobility Shift Assay) studies and then co-crystallized the enzyme with synthetic oligonucleotides containing bulky adducts (AAF-dG and dU-FI).

A replicator from tRNA with missing links to translation

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The replication of genomic information is central for the evolution of early life. However, long sequences are required to code for proteins and their replication requires the low error rate of proteins. This is a chicken-and-egg problem, often referred to as the Eigen paradox. To overcome it, life had to find a step-wise transition from archaic replication to modern translation, still driven by tRNA and ribosomes.

Here we present a replication mechanism solely based on tRNA. Its final structure readily allows to translate the succession of codon sequences into proteins. As starting material, double hairpins are formed from tRNA by five point mutations. They se-

lectively replicate a succession of anticodon sequences solely by hybridization in the temperature oscillation provided by thermal microconvection, possibly in rock pores of hydrothermal vents. The cross-catalytic and exponential replication takes less than five minutes and is confirmed with an explicit kinetic model. In the double hairpin conformation of tRNA, the amino acid binding sites are placed in close proximity to the anticodon sequence, allowing for specific interactions. The mechanism could have evolved from the selective degradation of single stranded over double stranded RNA. Was the RNA world rather a tRNA world?

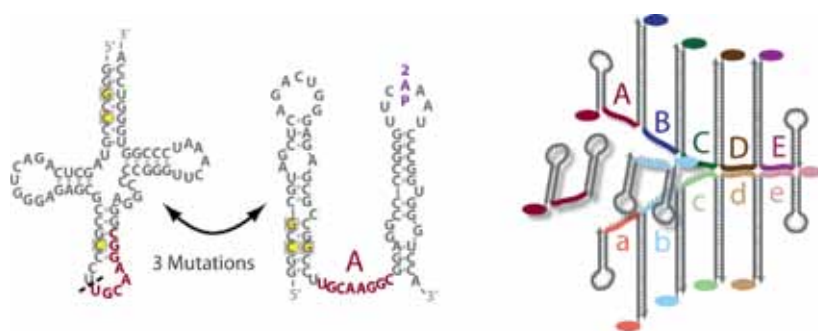


Figure 1: With only minor mutations, a tRNA from modern biology folds into a double-hairpin structure serving as fuel for an ancient information replicator. The information about a succession of sequence snippets, analog to modern anticodon regions, can be stored and replicated. The relationship to tRNA hints strongly towards a ancient translation mechanism.

Rigid molecular architectures for self assembly of gold nanoparticles

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Tuning light-matter interactions on a subwavelength scale allows the formation of materials exhibiting properties not existent in nature. The arrangement of metaatoms offers novel advances in fields like surface enhanced raman spectroscopy, solar cells, microscopy below the Abbe limit and cloaking devices. Fabrication of these materials can be realized by the formation of ordered structures with large electronic polarizabilities. Due to their tunable size, structure and optical properties as well as facile synthesis and stability gold nanoparticles are an excellent choice for these applications. Creation of supramolecular

architectures can be accomplished by linking of gold colloids facilitated through strong gold-sulfur interactions. Depending on the number of sulfur functionalities formation of dimers, trimers and larger clusters can be achieved. Here we report the synthesis of linear thio endfunctionalized oligo(p-phenylene) ethynyls and the selfassembly of gold nanoparticles mediated by these linkers.

Optical elements inside the vertebrate retina

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While cells are mostly transparent they are phase objects that differ in shape and refractive index. Any image that is projected through hundreds of micrometers of living tissue will normally be distorted by refraction, reflection, and scattering. Strangely, the retina of the vertebrate eye is inverted with respect to its optical function and light must pass through several tissue layers before reaching the photoreceptor cells.

In this work we focus on the optical properties of the photoreceptor nuclei that are stored in multiple layers directly before

the light sensitive segments. Based on micro-interferometry we show that a unique inversion of their spatial chromatin distribution in animals with a nocturnal lifestyle transforms these nuclei into micro-lenses. Computer simulations furthermore suggest that the arrangement of these nuclei in columns greatly improves transmission characteristics by a reduction of scattering and an effective channeling of light through the outer nuclear layer. This finding ascribes the first nuclear adaptation for an optical function and changes our understanding of the mammalian retina as an optical system.

[1] I. Solovei, M. Kreysing, Ch. Lanctot, S. Kosem, L. Peichl, Th. Cremer, J. Guck, and B. Joffe "Nuclear architecture of rod photoreceptor cells adapts to vision in mammalian evolution" *Cell* 137(2):356-368 (2009)

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Tuning the excitonic and plasmonic properties of copper chalcogenide nanocrystals

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Copper chalcogenides have recently been focus of interest, because of the need for semiconductor materials consisting of earth abundant elements for photovoltaic application,^[1] but more recently because of their unique structural and optical properties. Stoichiometric copper chalcogenides (Cu₂S/Se) exhibit a low intensity absorption onset up to 1000 nm (bandgap ~1.2eV) that increases in intensity towards the blue with a negligible absorption in the near infrared (NIR). In the presence of oxygen, their non-stoichiometric counterparts, Cu_{2-x}S and Cu_{2-x}Se, are formed with an absorption spectrum dominated by an intense band in the NIR. This has recently been assigned to free carrier absorption, i.e. plasmonic absorption.^[2-3]

In this contribution, we demonstrate the direct control over the evolution of the NIR plasmonic absorption upon oxidation and

reduction. These processes are correlated to the phase transition from stoichiometric to non-stoichiometric during the oxidation process, and back to stoichiometric upon reduction. This gives the unique opportunity to tune the position of the plasmon band in the near infrared. Moreover, we investigated the behavior of the excitons in Cu_{2-x}S NCs by controlling the evolution of the plasmon band. We report a quenching of exciton photoluminescence in the presence of the plasmon resonance which recovers upon reduction. Our results not only demonstrate that non-stoichiometric copper chalcogenide NCs simultaneously exhibit excitons and tunable localized surface plasmons in one material, but also that these materials are a unique platform to study at the nanoscale the interaction between excitons and free carriers.

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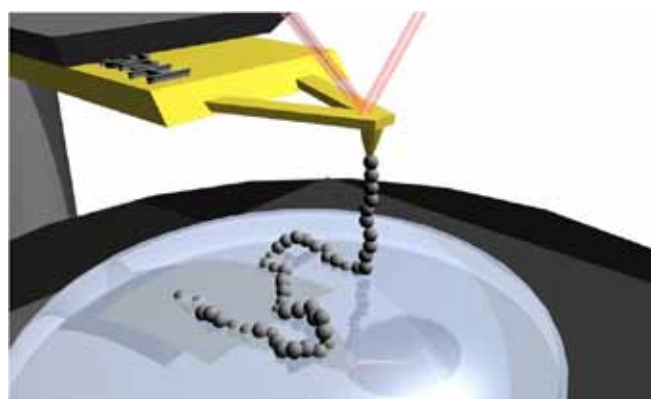
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Mechanism of single biopolymer adhesion

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The interaction of polymers with solid substrates, namely the adhesion of polymer molecules to a surface, is a central issue to numerous applications in materials science, engineering, biology and medicine. In this project the mechanism of adhesion between single biopolymers and solid substrates in liquid environment are investigated. During the last years AFM-based molecular force probes consisting of a single polymer covalently coupled to an AFM-tip were developed [1]. Such a single polymer can be adsorbed on solid substrates in aqueous environment several thousand times and the adhesion force determined by pulling them off the substrate. Usually plateaus of constant force are observed in the force extension traces, which yield important information on the influence of surface properties, pH or salt [2][3]. Possible information from the desorbed length is usually dismissed, because there is no clear picture on how variations in the desorption length report on molecular (non-equilibrium) interaction properties [4]. Here we present the characterization of the desorption length of a polymer in addition to the force and therefore gain qualitatively new information on adhesion of polymers at interfaces. In



particular, we study the temporal stability of the single polymer adhesion and how the desorbed length relates to the contour length for different adhesion forces.

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Large arrays of optical nanoantennas for single-molecule imaging and spectroscopy

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Nanochemistry Group, Chair for Photonics and Optoelectronics, Department of Physics and CeNS, Ludwig-Maximilians-Universität, Munich, Germany

When irradiated with light at the resonance frequency, the electric field between two gold nanoparticles in close proximity is greatly enhanced in a very confined, nanoscale volume, generating a plasmonic 'hot-spot'. A molecule brought in close proximity of this localized strong field can produce an enhanced optical signal such as raman scattering and fluorescence. In recent years, this concept of optical nanoantennas has attracted much attention as a superior approach for super-resolution imaging and spectroscopy.

Here, we introduce a method to generate large arrays of gold bow-tie shaped nanoantennas on solid substrates by bottom-up nanofabrication. The structure is generated by a combination of colloid lithography and subsequent plasma processing.

We are able to precisely control the tip-to-tip distance between billions of gold nanotriangles well below 20 nm which is superior to other methods reported so far and a prerequisite for optimum nanoantenna performance. In addition, the distance between adjacent gold triangles and the triangle size can be controlled independent from each other. We demonstrate the applicability of these plasmonic antenna arrays for fluorescence enhancement of molecules diffusing on a fluid supported lipid membrane and raman imaging spectroscopy. This implementation of near-field optics to a substrate on a truly macroscopic scale offers a broad range of applications for ultrasensitive optical detection and spectroscopy of single molecules well beyond the diffraction limit.

Building plasmonic nanostructures with DNA origami

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Metal nanostructures can support collective electronic oscillations at optical frequencies, known as surface plasmons (SPs). Existence of plasmons allows using subwavelength metal nanostructures to manipulate electromagnetic fields at the nanoscale. Moreover, the optical properties of such structures strongly depend on their geometry. This fact makes possible to engineer optical response of metal nanostructures over a broad range.

In this work we demonstrate DNA origami templated assembly of plasmonic nanostructures, specifically, assembly of helical complexes composed of metal nanoparticles. Such complexes show strong circular dichroism response in the visible wavelength range originating from the helical spatial arrangement of nanoparticles. To our best knowledge this is the first example of DNA origami templated assembly of plasmonic structures with engineered optical response.

Facing barriers: a precise modular nucleic acid carrier to investigate shielding, targeting and endosomolytic performance

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Pharmaceutical Biotechnology, Center for System-based Drug Research, and Center for NanoScience (CeNS), Ludwig-Maximilians-Universität Munich, D-81377 Munich, Germany

Non-viral polymer based conjugates are an attractive approach for drug delivery, since these compounds can be designed in a way to tune the carriers' properties regarding individual requirements. In case of nucleic acid delivery, several barriers have to be overcome within the delivery pathway, e.g. efficient nucleic acid complexation, prevention of unspecific interaction, specific uptake into target cells and intracellular delivery to the target site. A new solid-phase supported strategy for the sequential synthesis of poly(aminoamides) allows the assembly of precise tailor-made structures and the investigation of structure-activity-relationships. By this means a modular structure was assembled to investigate shielding, targeting and endo-

somal escape performance of the delivery devices. The structure is built up of a poly(aminoamide) backbone capable of nucleic acid complexation together with a shielding poly(ethylene glycole) and targeting ligand attached to a defined position. The endosomal escape can be enhanced by introduction of several histidine residues in order to increase the 'proton sponge effect' or by use of endosomolytic siRNA. The model shows the opportunity to investigate the effect of the different modules and to suit the carriers' properties to the separate delivery steps and biological barriers.

Dynamic photonic crystal membranes based on responsive polyelectrolyte gels

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In the last decade, photonic crystals (PhCs) in combination with "smart"/ stimuli-responsive materials (especially hydrogels) have been demonstrated to be a class of attractive optical transducers for sensing due to their high chemical sensitivity, availability, and straightforward/unsophisticated fabrication. Analytes capable of giving rise to a change in either the refractive index or the structural periodicity of the PhC can affect its Bragg diffraction and therefore the optical spectra the PhC displays, which allows non-destructive, label-free detection. Here we report on a new class of 2-D PhC-based sensors - dynamic photonic crystal membranes (DPCMs). Unlike traditional PCMs,

which are unexceptionally rigid, the DPCMs are made from responsive polyelectrolyte gels and allow large volume changes when exposed to stimuli and hence, an enhanced optical readout. We demonstrate that such PCMs offer intrinsic guided modes and Fabry-Pérot interference, and that both can therefore be exploited for optical transduction. The DPCMs have shown an ultrafast response due to their submicron thickness. A colorimetric readout can be readily achieved thanks to the considerable change of the DPCMs' thickness in response to various external stimuli.

Macroporous indium tin oxide (ITO) films as nanostructured transparent electrodes for organic photovoltaic devices

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Organic photovoltaic devices attract tremendous attention as potential sources of renewable energy. Their efficient performance requires an effective collection of photo-generated charges and their fast transport to the current collectors. One of the possible ways to achieve this goal is to integrate the nanostructured transparent electrodes into the active material, which could provide the efficient collection and transport of charge due to their large and accessible surface area and high conductivity. In order to prove this concept, we have fabricated polymer solar cells based on the nanostructured transparent indium tin oxide (ITO) electrodes instead of the commonly used flat ITO films (Figure 1). We have developed a strategy to elaboration of macroporous ITO layers via co-assembly of poly-methylmetacrylate (PMMA) spheres with a diameter of ca. 370 nm and preformed indium tin hydroxide nanoparticles. By

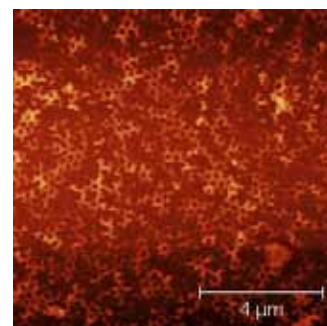
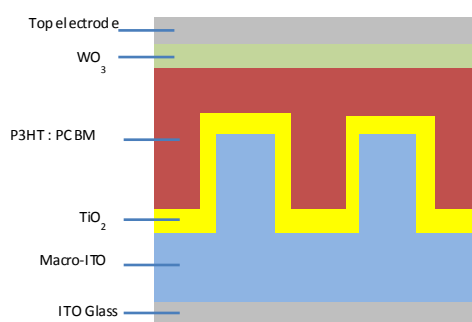


Figure 1: (a) Scheme of the organic solar cell with the nanostructured ITO electrode. (b) AFM image of macroporous ITO layer.

this way, periodic porous structure with uniform accessible pores with the size of ca. 250 nm and are obtained which were used for fabrication of inverted organic solar cells.

Assembly of a multi-color TIRF setup

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² Department of Physics, University of Illinois at Urbana-Champaign

Biophysical processes at the cellular level are best understood when no ensemble average distorts the overall picture, but rather single molecules are investigated sequentially.

Foerster or Fluorescence resonance energy transfer (FRET) measured between a single donor fluorophore and a single acceptor fluorophore is an ideally suited technique to measure distances and dynamics at single-molecule level. Though single-pair FRET is a powerful tool to study biological molecules undergoing Intra- and Inter-molecular interactions, it is restricted to only one measurable distance per molecule of interest. Owing to this limitation, it cannot reveal intricate details of the global conforma-

tional changes during complex biomolecular interactions which are multi-dimensional. For large complexes, using multi-color FRET schemes helps reporting about conformational changes in separate domains of the macromolecule in real time and provides a more comprehensive picture.

In order to expand the single-molecule FRET to study complex molecular interactions, we have assembled a Four-Color Total Internal Reflection Fluorescence (TIRF) Microscope system which allows to measure distances between four fluorophores and thus gives insight into complex molecular dynamics.

Optoelectronic properties of TiO₂ lamellas fabricated by focused ion beam lithography

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We present studies towards revealing a correlation between the nano-morphology of optoelectronic devices fabricated by a focused ion beam (FIB) lithography and their optoelectronic properties. For the device fabrication, first a thin film of TiO₂ with a sponge-type morphology is prepared by a sol-gel process on a glass substrate [1]. Then, a carbon protective layer (thickness: ~2 μm) is deposited by an ion beam induced deposition (ibid), and in turn, a lamella is cut out by FIB-milling. The lamella is picked up with the help of a micro-manipulator, and it is transferred to an electronic circuit with two Au electrodes with a gap of ~10 μm. On both sides of the lamella, Pt contacts are

deposited by an ibid-process for electronically contacting the TiO₂ film. The cross-section characteristics of the lamellas are investigated by a spatially resolved optoelectronic technique. We observe the effects of Schottky contacts, trap state dynamics, indications of a bolometric photoresponse, and a photoconductance originating from a multi-photon absorption process.

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Ionic Liquids as alternative imaging media for self-assembled nanostructures by (cryo-)TEM

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With the development of modern polymerization techniques in the last decades an increased control over the molecular structure of polymers by means of composition, topology and functionality is achieved. Through this, defined polymer structures with tunable self-assembly in the nanometer dimension, such as into micelles, vesicles and worm-like structures, became available. Due to the resulting growing need for characterization, electron microscopy became a powerful tool in polymer science caused by the nm-scale resolution and the visualization of individual structures in contrast to, e.g., dynamic light scattering. In this poster recent examples of the investigation of supramolecular aggregates by cryo-TEM will be presented. However, cryo-TEM is mostly restricted to water-based solutions and its compatible copolymer systems. Furthermore, low contrast for most of organic polymers often requires an additional staining procedure, which is poorly controllable and impairs the formation of vitrified ice films. Recent studies were conducted to elucidate alternative preparation approaches to improve the contrast generation in self-assembled supramolecular micelles, thus allowing the structural characterization of the different constituents of the structures (Fig. 1b), by the utilization of ionic liquids (IL). They permit (i) tunable physical properties (e.g. polarity, hydrophilicity) by different cation/

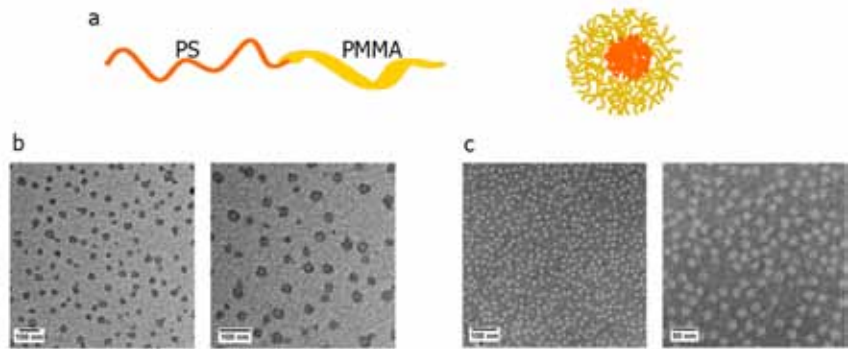


Figure 1: Investigation of PS-PMMA micelles (a) assembled in an ionic liquid. b) Detection of the shell thickness: The micelle/IL solution was immobilized on a grid and washed with acetonitrile, whereas the IL remains only in the loosely packed PMMA-block. c) cryo-TEM of the micelles in IL shows the PS-core with inverse contrast

anion combination allowing for assembly of a wider range of polymer systems, while forming stable films under common cryo-vitrification procedure; (ii) the intrinsic staining of the IL by the anion prevents staining procedures while enhances the contrast (Fig. 1c). In addition, due to their low vapour pressure in a wide range of temperature and their distinct surface tension – which allows the formation of stable free-standing liquid support films – ionic liquids were also used for the investigation of different block copolymer assembly by room temperature TEM.

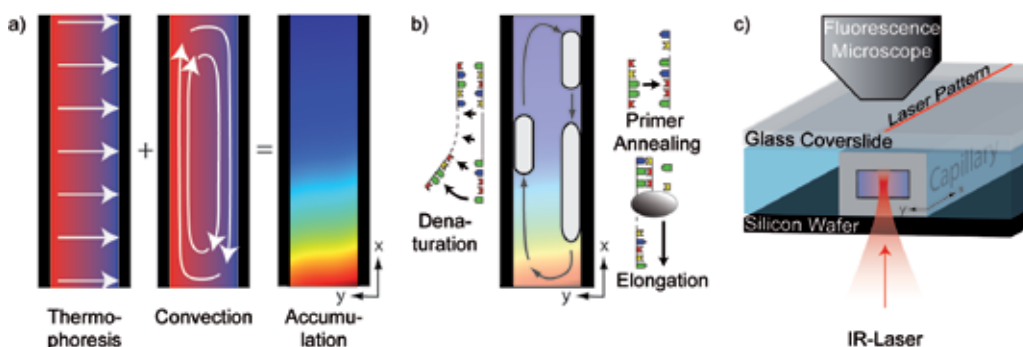
A thermal trap for DNA replication and polymerization

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Fakultät für Physik und Center for NanoScience (CeNS), Ludwig-Maximilians-Universität, Munich, Germany

Normally, genetic information is protected from free diffusion into the environment by cell walls. The cells provide all conditions for the replication and mutation of genetic material - the basic prerequisite for Darwinian evolution. We modeled a fundamental principle in the laboratory, which allows for continuous evolution of genetic material without cell membranes. We filled a thin capillary with DNA and nucleotides, solved in buffer as an aqueous solution (Figure c). A moving infrared laser spot then generated a thermal gradient realizing thermophoresis as well as a cyclic convection of the solution. The superposition of both effects caused the double-stranded DNA molecules to

migrate to the cold area (Figure a), while simultaneously cycling the DNA between the cold and warm section of the capillary. In the hot area, the DNA separates into single strands. These are then elongated by a polymerase enzyme in the cold region to two double-stranded copies of the original template DNA (Figure b). Therefore, a simple temperature gradient drives both, an exponential replication as well as the selective accumulation of information. This is relevant as presumably similar thermal conditions prevailed in rock pores near hot undersea springs of prehistoric oceans. Our experiment shows how a simple disequilibrium setting may allow life to evolve. Furthermore,



we are interested in the combination of polymerization/degradation and thermal trapping of DNA/RNA. Preliminary results show a massive boost of accumulation yielding towards a double exponential selection and creation of longer polymers compared to shorter ones.

Thermodynamics and kinetics of cruciform extrusion in a coarse-grained DNA model

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¹ Rudolf Peierls Centre for Theoretical Physics, University of Oxford, United Kingdom

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³ Physical and Theoretical Chemistry Laboratory, University of Oxford, United Kingdom

Cruciforms are noncanonical conformations of DNA that can occur in strands whose sequence exhibits an inverted repeat (IR) symmetry (Fig. 1). This permits the formation of intra-strand bonds as hairpins situated opposite to each other along the double-stranded B-helical DNA. Cruciform structures are favoured in the presence of undertwist, where they provide a means to relax torsional stress. They have been implied to be relevant in a number of essential cellular processes, including gene regulation and DNA repair [1]. We use a coarse-grained model of DNA developed by Ouldridge et al. [2] in order to study free energy landscapes and determine extrusion kinetics of cruciform formation at different temperatures and degrees of supercoiling (Fig. 2). We find a rapid transition behaviour between the B-helical and cruciform states relative to the lifetime of the respective conformation. Cruciform hairpins are found to nucleate in a correlated, but not synchronous way, providing a more detailed picture of the S-type kinetics inferred from bulk

thermodynamic experiments such as 2d gel electrophoresis. Our findings are consistent with recent single molecule probes of cruciform extrusion [3].

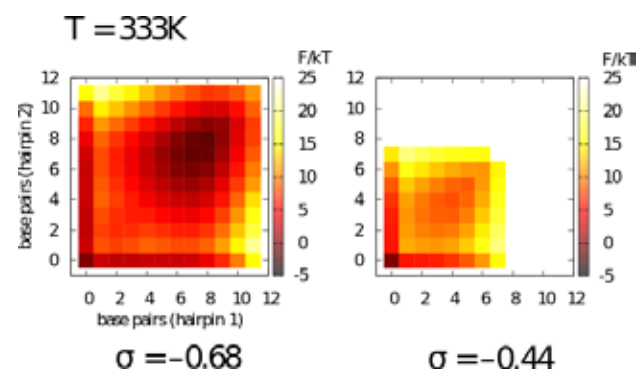


Figure 2: Free energy landscapes of cruciform extrusion at $T = 333\text{K}$ and superhelical densities $\sigma = -0.68$ (left) and $\sigma = -0.44$ (right).

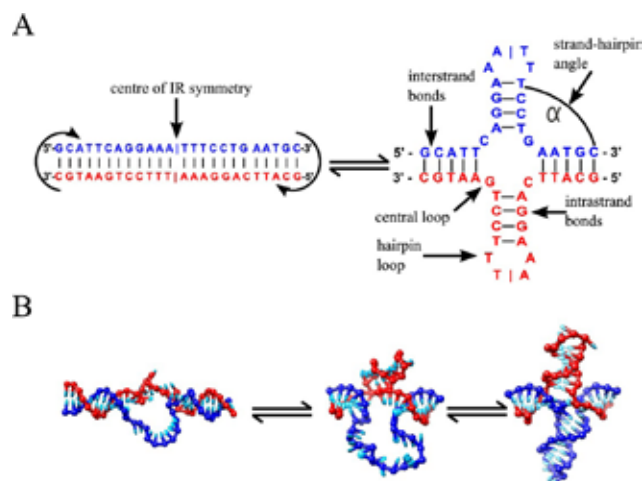


Figure 1: (A) Schematic of cruciform formation in a DNA strand exhibiting a palindromic sequence. (B) Typical bubble, single arm and fully formed cruciform states from simulations.

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Targeted delivery of gene vectors into cancer cells

F. M. Mickler, Y. Vachutinsky, M. Oba, N. Ruthardt, E. Wagner, M. Ogris, K. Kataoka, C. Bräuchle

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Optical lithography techniques are widely used for fabrication of nanoscale devices. The ability to fabricate such structures with arbitrary size and shape is essential for their wide applications in optoelectronics, biological and medical sciences. The conventional far-field lithography is a diffraction limited technique which is not suitable for structures with lateral feature size beyond the diffraction limit. Some far-field lithography techniques can go beyond this limit, but they do not allow arbitrary pattern formation. Scanning near-field optical microscopy techniques can also be used for nanostructure production beyond the diffraction limit. However, they are characterized by low throughput, due to their serial scanning nature and pose

restriction for maximal structure size. Here we introduce a new optical lithography technique, optical force stamping lithography (OFSL). This approach employs optical forces on single nanoparticles in multiple focused Gaussian beams produced by a spatial light modulator. The so formed optical stamp provides rapid immobilization of single nanoparticles onto a substrate with positioning accuracy well beyond the diffraction limit. The optical stamp can be changed tens of times per second due to the optical nature of the process. OFSL is not restricted to nanoparticle type or substrate. We believe that these evident advantages of the optical force stamping lithography will make it a standard tool for fabrication of nanodevices.

Bioaffinity separations employing thermally-responsive polymers

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We present an integrated bioseparation and detection system comprising thermally-responsive nanoparticle mixtures for point-of-care diagnostic immunochromatography. The system allows for labeling of the target biomarker with colloidal gold, followed by magnetic separation and enrichment. Detection by lateral flow immunoassay is used to visualize the biomarker target at the test line of a nitrocellulose flow strip. The nanoparticle system is composed of a mixture of iron oxide and gold nanoparticles, each with a thermally-responsive poly(N-isopropylacrylamide) (pNIPAm) coating. These “smart” polymers undergo a hydrophilic to hydrophobic phase transition upon heating above the lower critical solution temperature (LCST) of ~32 degrees celsius. Upon raising the temperature, the two particle types co-aggregate, forming magnetic/gold agglomerates. Subsequent application of a magnetic field results in magnetophoresis and separation of the biomarker bound to the gold nanoparticles (AuNPs). Enrichment is achieved upon discarding the supernatant, and dissolving the captured aggregates into a smaller volume of cold carrier fluid. Detection of the enriched gold-labeled biomarker by immunochromatography is shown to be more sensitive than conventional non-concentrating approaches.

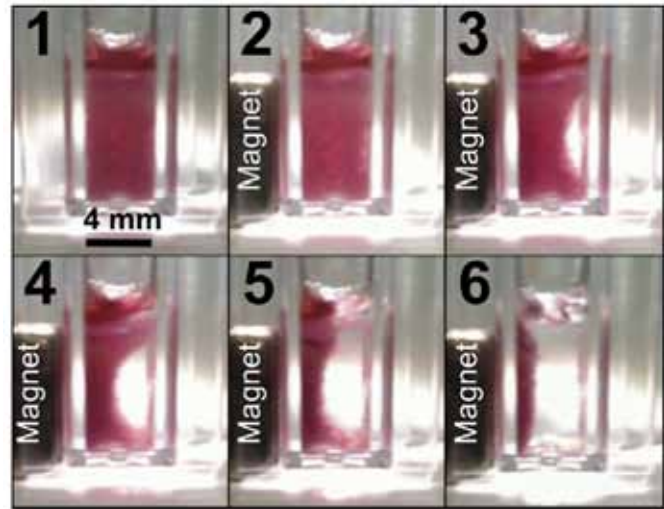


Figure 1: Time-lapse images showing magnetic separation of AuNPs. (Image 1) A temperature stimulus is applied to the AuNP/ mNP mixture, causing polymer phase transition. (Images 2-6) A magnetic field is applied and the AuNPs are pulled to the side of the cuvette via polymer-induced co-aggregation with mNPs. Capture time=30 min.

Optical force stamping lithography

Spas Nedev, Alexander S. Urban, Andrey A. Lutich and Jochen Feldmann

Photonics and Optoelectronics Group, Physics Department and CeNS, Ludwig-Maximilians-Universität München, 80799 Munich, Germany

Optical lithography techniques are widely used for fabrication of nanoscale devices. The ability to fabricate such structures with arbitrary size and shape is essential for their wide applications in optoelectronics, biological and medical sciences. The conventional far-field lithography is a diffraction limited technique which is not suitable for structures with lateral feature size beyond the diffraction limit. Some far-field lithography techniques can go beyond this limit, but they do not allow arbitrary pattern formation. Scanning near-field optical microscopy techniques can also be used for nanostructure production beyond the diffraction limit. However, they are characterized by low throughput, due to their serial scanning nature and pose

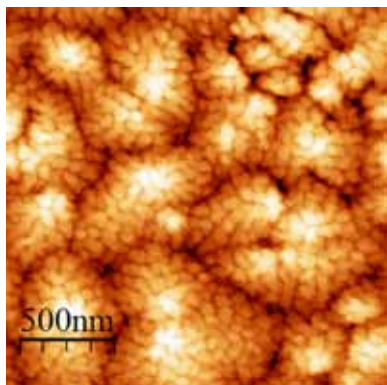
restriction for maximal structure size. Here we introduce a new optical lithography technique, optical force stamping lithography (OFSL). This approach employs optical forces on single nanoparticles in multiple focused Gaussian beams produced by a spatial light modulator. The so formed optical stamp provides rapid immobilization of single nanoparticles onto a substrate with positioning accuracy well beyond the diffraction limit. The optical stamp can be changed tens of times per second due to the optical nature of the process. OFSL is not restricted to nanoparticle type or substrate. We believe that these evident advantages of the optical force stamping lithography will make it a standard tool for fabrication of nanodevices.

In-situ characterization of the formation of a pentacene / C60 heterojunction in an ambipolar organics thin film transistor: thickness resolved transport

Simon Noever, Bert Nickel

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Organic-organic semiconductor heterojunctions have become subject to intense research over the last few years due to their importance for plastic electronic devices such as organic solar cells, organic light emitting diodes and light emitting thin-film transistors (TFTs). In contrast to regular, inorganic p-n junctions, the situation of the level of doping and the density of interface states in organic junctions is unclear. The active materials are often chosen to work as either p-type or n-type semiconductors based on their band alignment to the electrode work functions and their intrinsic charge carrier mobilities.



Atomic force microscopy high image of a 20nm pentacene / 25nm C60 dual layer, grown on a cyclic olefin copolymer finished silicon nitride substrate.

Pentacene and C60 show the highest mobilities for holes and electrons in organic thin films, respectively. Here we study the pentacene / fullerene C60 heterojunction. Structural analysis of the system on silicon nitride (Si_3N_4) substrates via AFM and X-ray reflectometry show a correlated growth of C60 on pentacene and good crystallinity of the organic semiconducting layers.

For the thickness resolved experiment we first assembled a bottom-gate, top contact pentacene transistor on a COC (cyclic olefin copolymer) finished Si_3N_4 dielectric. While growing the C60 layer on top of the unipolar TFT by vapour deposition, the I-V curves of the device are measured constantly. Thus we gain information on the fullerene thickness dependent characteristics, namely the mobilities, threshold voltages and hysteresis effects.

A key result is the threshold shift of the p-channel to more positive values once the n-channel opens. This bias suggests the heterojunction to act as an additional top-gate. The thickness information of the junction combined with the electrical characteristics of the transistor represent a basis for spatial trap and channel models for organic heterojunction setups, complementing established methods such as UPS and XPS.

Molecular games using interacting replicators

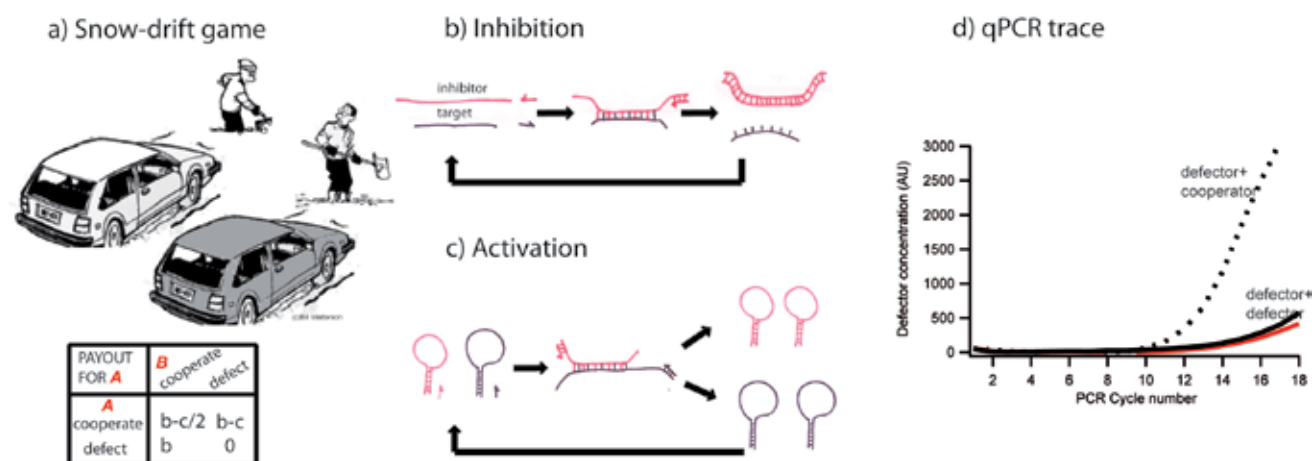
Natan Osterman, Franziska Kriegel, Erwin Frey and Dieter Braun

Fakultät für Physik and Center for NanoScience (CeNS), LMU München, Germany

Game theory was used to model the interactions of humans and animals. Recently, game theory was also applied to the behavior of yeast and bacteria. As games are played at smaller and smaller scales, which is the most minimal level at which games can be played? Here we show that replicating molecules implement the standard games of game theory (e.g. snowdrift game, Figure a). The replicator sequence determines the molecule's strategy by means of mutual and intramolecular binding interactions. In RNA, such interactions are typically strong and cannot be easily overcome by for example temperature cycling. For competing replicators in an RNA-world scenario, molecular games are likely to play a prominent role. We explore the possible cooperating interactions to replicate the fastest. We implement replication by the DNA-based Polymerase Chain Reaction (PCR). Depending on replicator sequences they can

either inhibit or activate each other or have no interaction at all. Inhibition is achieved when primer binding region of one replicator ("target") is blocked by competing replicator ("inhibitor") (Figure b). As a result the "target" growth is much slower. Activation plays a prominent role in the replication of hairpins. Such molecular configuration is self-blocking its primer region and therefore can't be replicated. However, if another molecule ("activator") binds to the hairpin loop, the loop is opened and the molecule is free to replicate (Figure c).

The exponential growth phase is used to derive game dependent fitness and payout matrices (Figure d). Interestingly, a change of the boundary conditions let identical molecules to switch between for example by-product mutualism and the snowdrift game.

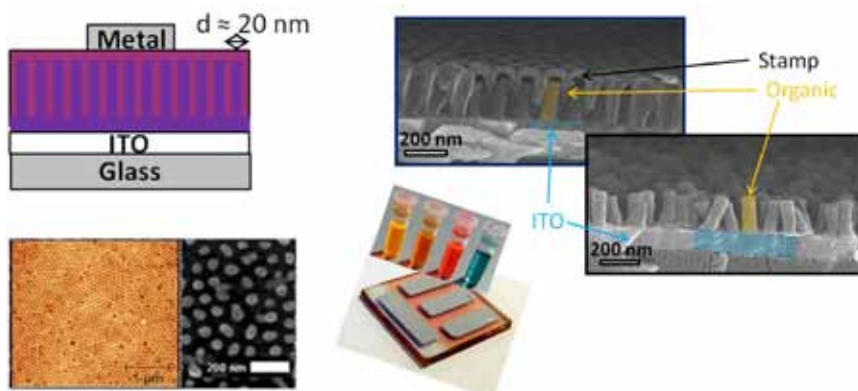


Nanostructuring donor-acceptor interfaces in organic photovoltaic devices

Claudia M. Palumbiny, Sarah Lindner, Holger C. Hesse and Lukas Schmidt-Mende

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Precise control of the heterojunction morphology is one of the key issues for the improvement of organic photovoltaic devices.[1] Interdigitated interfaces of the organic materials on the nanometer length scale allow for comprehensive exciton harvesting in the active layer and provide ideal charge extraction pathways for separated charge carriers towards the electrodes. Thus increased current generation and reduced bimolecular recombination can be accessed in the organic photovoltaic devices (OPV). Using anodized aluminum oxide (AAO) hard templates as stamps nanostructured organic layers can be realized on ITO support. Precise control of the anodization process facilitates the control of the template dimensions including pore length, pore to pore distance and diameter independently. As shown recently, a direct replica of the template structures can be transferred into the organic compounds using thermal and photo-induced polymerization processes.[2] Experience in fabrication techniques including solution processing and vacuum sublimation techniques [3] as well as different device geometries [4]



enables us to study the device physics of these novel architectures in OPV devices. Exciton generation and recombination are studied by spectroscopic measurements such as photoluminescence analysis and transient absorption spectroscopy. Intensity and temperature dependent measurements on the devices will provide insight to charge carrier trapping and help to quantify the bimolecular recombination. Furthermore, imprinting induced changes on the molecular alignment can be probed by x-ray analysis.

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Mechanochemically induced hydrolysis of acyloxysilanes

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The mechanical degradation of macroscopic polymeric materials is directly related to the rupture of individual chemical bonds, which determine the properties of synthetic and natural materials.¹ To obtain a deeper understanding of material failure it is essential to understand the breakdown of individual chemical bonds.

We investigated the temperature-dependent lifetime of individual chemical bonds as a function of the clamp force at pH 2.0 using an atomic force microscope (AFM) in the force spectroscopy mode. Single molecules of carboxymethylated amylose (CMA) were tethered between a silanized glass substrate and a silanized silicon nitride AFM tip via acid-catalyzed ester con-

densation forming an acyloxysilane.² Individual CMA molecules were then stretched until the clamp force was reached, and the force was kept constant until a spontaneous bond scission was observed.

Reaction kinetics was followed at varying temperature and clamp force exhibiting a bi-exponential behaviour. Data analysis with two alternative models both based on Arrhenius kinetics point to the hydrolysis of the acyloxysilane in the surface anchor depending on the residues at the silicon.³ This was corroborated by first quantum chemical calculations indicating that the reaction kinetic of the Si-O bond rupture in the acyloxysilane is considerably affected by the side groups at the silicon atom.

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Single molecule cut and paste for protein based functional assembly

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Single-molecule cut-and-paste surface assembly (SMCP) has formerly been employed in the controlled deposition of individual fluorophores in well-defined nanometer sized patterns [1]. The technique allows for the creation of patterns of arbitrary shape and with arbitrary numbers of single molecules consisting of multiple species. The accuracy has been shown to be ± 10 nm with the given spacer and DNA sequence lengths [2]. SMCP has been also used to build up a Biotin scaffold that Streptavidin-coated nanoparticles could bind to [3]. Utilizing specific molecular interactions, for example between DNA-binding proteins and DNA or antibodies and antigens, this technique is capable of providing a scaffold for the controlled self-assembly of functional complexes. Furthermore, this allows for the introduction of SMCP into protein science. We aim to employ DNA-binding Zinc-finger variants and GFP-binding nanobodies as shuttle-tags fused to the proteins of interest. Thus a fully expressible system that can be used for the step-wise assembly of individual building blocks to form, for example, large enzyme complexes or protein networks, is provided.

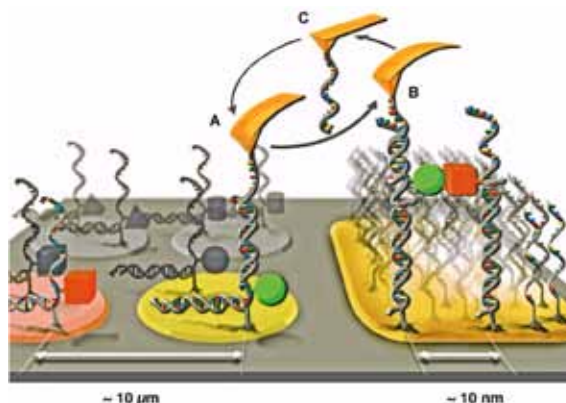


Figure 1: Cartoon of a DNA-based single-molecule cut-and-paste process. Length and binding geometry of the oligomer handles are chosen so that a hierarchy of unbinding forces allows the repetition of this process over and over again [1].

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Quantitative analysis of bacterial interaction leading to pattern formation

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Bacterial communities represent complex and dynamic ecological systems. Different environmental conditions as well as bacterial interactions have determining influence on establishment and conservation of bacterial diversity and can lead to so-called pattern formation. Stable coexistence of several bacterial strains is often only possible under well-defined conditions.

To study the development of bacterial populations we use time-lapse microscopy to investigate the colicin E2 system of three *Escherichia coli* strains labeled with different fluorescent proteins. Combinations of these strains, with distinct growth

parameters, lead to either instable, metastable or stable coexistence. Besides growth rate and colicin production, coexistence was mainly influenced by lag time variations. In accordance with the results, two main strategies lead to survival: sensitive strains need short lag phases and rapid growth rates, while toxin producing strains even with extended lag phases and slower growth rates can prevail.

Specific growth parameters enable cyclic dominance, where the colicin-producing strain kills the sensitive strain, outgrows the resistant one. This in turn has a growth advantage over the first.

Aggregation and polydispersity of lipid coated, colloidal, mesoporous silica nanoparticles

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Fluorescence correlation spectroscopy (FCS) is an optimal technique for exploring diffusion dynamics and interaction of nanoparticles. Previous studies have shown that colloidal mesoporous, silica (CMS) nanoparticles [1] show a great potential as versatile multifunctional drug delivery system for selective targeting of malignant cells [2]. Recently we showed that lipid bilayer encapsulated CMS nanoparticles can be achieved using a solvent exchange method to fuse lipids and nanoparticles [3]. However, for biomedical applications a fine control size distribution and surface functionalization of nanoparticles is important to achieve optimal biodistribution and to avoid blood clotting. Here we investigate the size distribution and aggrega-

tion behavior of CMS nanoparticles compared to lipid bilayer coated CMS nanoparticles as well as study their interaction with proteins (transferrin, VWF). We use FCS with one- and two-photon excitation and electron microscopy to characterize the nanoparticles. The home-build two-photon FCS setup is dedicated to studies in complex media like blood plasma and tissue. Two-color cross correlation measurements of the lipids and the nanoparticles allow to determine binding. We find a wide range (20nm-200nm) of CMS nanoparticle-diameters as well as an intrinsic polymorphism depending on nanoparticles synthesis. After lipid deposition cross correlation measurements show a ratio of more than 60% lipid coated nanoparticles.

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Colloidal porous frameworks for the fabrication of photonic multilayer systems

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Zeolitic imidazolate frameworks (ZIF) are a class of microporous compounds in which metal atoms such as zinc are connected through imidazolate ligands to form rigid frameworks. Due to their tailorable porosity, ZIFs represent attractive candidates for gas separation or sensing applications.[1, 2] Recently, prototypical ZIF-8 has been described as a selective sensing material for gases and vapors.[3,4] According to Lu and Hupp, the infiltration of guest species in ZIF 8 thin films can be monitored by the change of the effective refractive index. [4] Theoretically, this detection method can be transferred to photonic multilayer systems by stacking alternately thin films

of different dielectrics such as ZIF-8 and TiO₂, yielding 1D photonic crystals dubbed Bragg stacks. In principle, the resulting interference colour of the Bragg reflector can be tuned by the ad- and desorption of guest species such that the multilayer system can be used as an optical sensing device.[5] By successively spin-coating colloidal suspensions of TiO₂ and ZIF-8, we were able to produce functional photonic multilayer stacks.[6] Optical sensing measurements have recently shown that the infiltration of guest species leads to spectral changes which are specific for particular analytes.[6]

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Collective motion of cooperating molecular spiders

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Molecular motors are known to carry cargo along tracks. While ubiquitous in nature, it is still a challenge to construct molecular motors synthetically. Recent theoretical [1] and experimental [2] studies suggest that so-called molecular spiders might act as molecular motors. Molecular spiders consist of an inert body and several legs which walk on a lattice. The legs cleave the substrate on the lattice which enhances the hopping rates. For 1-dimensional tracks, single spiders experience a bias towards uncleaved sites. However, this bias is rather small. Motivated from cooperative mechanisms of molecular motors, we consider a minimal model in which molecular spiders display collective behaviour. This leads to cooperative transport of cargo on a 1-dimensional track. Using analytical calculus and extensive stochastic simulations, we found that two or more cooperating spiders show better transport properties compared to single spiders. In particular, the mean traveled distance is increased significantly for cooperative spiders. Likewise, the spatial distribution of the spiders is sharpened, what reduces the noise. We expect that these results on cooperative behaviour may hold for other classes of molecular motors as well.

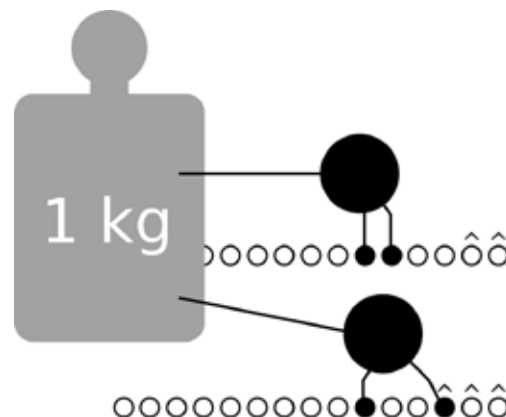


Figure 1: Two molecular spiders pulling a cargo. Hats indicate lattice sites with substrate.

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Nonadiabatic dynamics of two strongly coupled nanomechanical resonator modes

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Landau-Zener transitions^{1,2,3,4} play an important role in numerous quantum phenomena and have been investigated both experimentally and theoretically in many fields of physics. We present a single nanomechanical resonator as a classical model system with which such a dynamical behaviour can be realised and studied.

Our experiments are performed on a doubly clamped high stress silicon nitride beam which is dielectrically coupled to a $\lambda/4$ microwave cavity to enhance signal sensitivity for the detection of both the in- and out-of-plane modes of the resonator. It also allows for the tuning of the respective resonance frequency of both modes via the incident microwave power.

When changing the detuning between the fundamental in- and out-of-plane mode we observe a characteristic anticrossing re-

flecting strong coupling between the two modes. The model of two linearly coupled harmonic oscillators is fitted to the data with excellent agreement in order to extract the coupling parameters of our system.

To explore the dynamic behaviour we perform pulsed measurements. Here, the system is initially driven in one of the branches of the spectrum, then the detuning is swept through the anti-crossing region at different speeds and the oscillation amplitude in each of the branches after passage through minimal detuning is monitored. For faster sweep rates, the probability of transferring energy between the low and high energy branch (diabatic transition) increases, whereas it becomes less likely to remain within the same branch (adiabatic transition). Thus our findings are well described by the model of a classical Landau-Zener transition.

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Highly oriented mesoporous silica channels synthesized in microgrooves and visualized with single molecule diffusion

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Mesoporous silica materials play an important role in host-guest chemistry. Their well-defined periodic pore system and their enormous flexibility regarding functionalization enable them to accommodate a variety of different guest molecules. For many applications it is highly desirable to have large ordered domains of channels and a high mobility of guest molecules inside the latter. Here we describe the synthesis and characterization of mesoporous silica thin films with large, well-aligned mesopores.[1] The alignment was achieved by combining an evaporation-induced self-assembly (EISA) approach with guided growth inside poly(dimethylsiloxane) (PDMS) microgrooves. The characterization and visualization of the mesoporous network was done with single-molecule fluorescence microscopy. Fluorescent dyes that are incorporated into the mesochannels during synthesis can be used as probes to map the internal structure of the pore system in the as-synthesized silica films and provide direct feedback for tuning synthesis condi-

tions (see Figure 1). Additionally, high-accuracy single molecule tracking experiments provided even deeper insight into the real nanoscale structure of the silica host. The high spatial resolution of this technique showed that the guest molecules diffuse in a 1D-random walk-like manner along the highly-oriented parallel channels and occasionally switch from one channel to another through small defects in the pore walls. These findings demonstrate that imperfections which may block the diffusional pathway of molecules may be counterbalanced by defects in the side-walls allowing the molecule to circumvent dead ends. Present studies also address issues such as the influence of electric fields on charged guest molecules (e.g., charged dyes or biomolecules) inside a porous silica host.

Financial support by SFB 749 and the cluster of excellence Nanosystems Initiative Munich (NIM) is gratefully acknowledged.

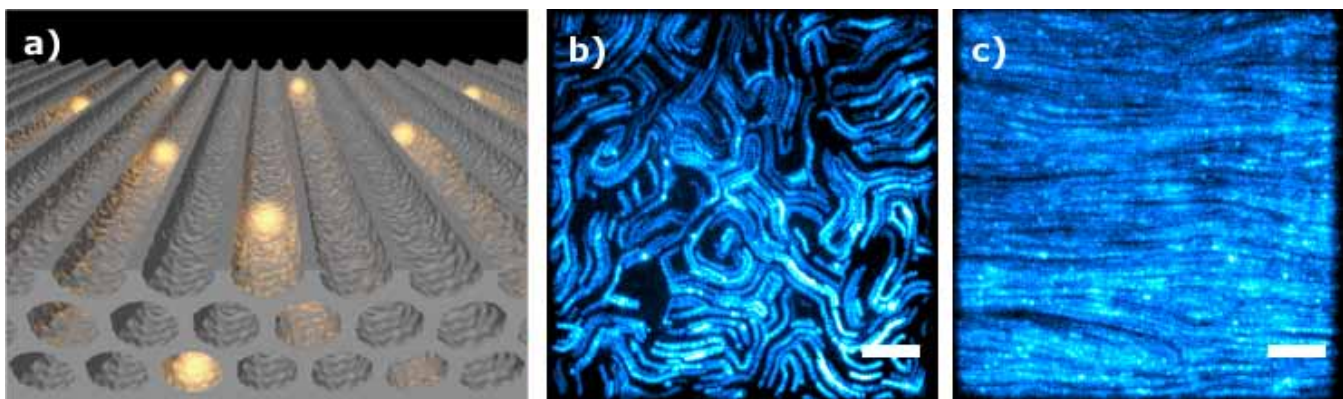


Figure 1: a) Schematic representation and b,c) “maps” of the different porous silica hosts obtained with single molecule microscopy (false-colored in blue; scale bars are 5 μm).

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Wave localization and focussing in bistable systems

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Bistable systems arise in a broad range of fields in biology, chemistry and physics. Such systems are often subject to external gradients, which may be morphogen gradients in cell biology or temperature gradients in ecology. In many scenarios these inhomogeneties allow for the localization and focussing of propagating fronts. We here study a biologically motivated class of models with different types of external gradients. Our analytical calculations reveal the complete phase diagram of pa-

rameters, which allow wave localization. Beyond that localized waves are subject to external perturbations and internal noise. We investigate the optimal conditions which minimize those sources of defocussing. Our results have possible implications for bio-technology, population dynamics and cell biology.

Solid-phase supported synthesis of precise branched polymers for gene delivery

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Missing or defective genes are the cause of multiple severe diseases like various cancer forms. Therefore, scientists working in the field of gene therapy have been striving for decades to find a safe and efficient gene carrier system. This quest emerged due to the inability of “naked” genes to overcome the biological barriers associated with systemic or local delivery on their own. Among others, cationic polymers like polyethylenimine have the ability to reversibly compact pDNA through electrostatic interaction into stable polyplexes. Thus, the payload can reach undamaged the target cell compartment, namely the nucleus, where it can fulfil its task. Such polymers, although showing promising results in terms of transfection efficiency and cytotoxicity, are still accompanied by a crucial drawback which hampers essential structure-activity relationship studies: polydispersity. The concept of solid-phase supported synthesis (SPS) shows a way out of this dilemma. This method allows step-by-step polymer chain elongation and therefore exact position control of the inserted compound.

Hence, we present here two classes of precise branched cationic polymers for gene delivery consisting of cationic building blocks and amino acids:

1) Branched non-biodegradable polymers were directly assembled on solid phase, implying the advantage that with only few synthetic steps a higher molecular weight (Mw) can be achieved, which is required for efficient gene delivery.

2) Linear chains synthesized on solid phase were used for the modification of an activated precise dendritic core via disulfide bond formation. The resulting biodegradable product dissociates in the cytosol due to the reducing environment, thus preventing cytotoxicity related to high Mw.

Still, this is not the end of the story, as SPS offers a broad spectrum of possible site-controlled modifications, encouraging the hopes for very efficient and well tolerated gene carriers.

Integrin generated forces on prostate cancer bone metastasis

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Integrins are heterodimeric cell receptors which take part in cell attachment processes between a cell and the tissue surrounding it, which may be other cells or the ECM. Since the most abundant extracellular matrix protein in bone are the collagen type-I proteins, collagen receptor integrins ($\alpha 1\beta 1$, $\alpha 2\beta 1$ and $\alpha 11\beta 1$) may have an important role in attachment of prostate cancer (PC) cells to bone tissue and development of cancer metastasis. In this study we have used atomic force microscopy (AFM) to identify cell adhesion forces generated by collagen receptor Integrins. The measurements were performed with PC Cells (PC3-Bone metasis Type, LnCAP-Lymph node metastasis

type) and bone residing mesenchymal stem cells (SCP1). The effects of integrin generated forces on cell to matrix (collagen type-I) and cell to cell (PCs to MSCs) adhesions are investigated. According to our AFM results, we found that PC3 cells adhere with a higher rate, higher number of binding steps and stronger binding forces than LnCAP cells to collagen matrix proteins and mesenchymal stem cells. Moreover our gene expression studies showed that Collagen receptor integrins expression level are much lower for LnCAP than PC3. Collagen receptor integrins seem to have an important effect on cell adhesion to ECM and other cells in PC metastasis in bone marrow.

How to open a ligand gated ion channel by the Patch-Clamp-AFM

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Ligand gated ion channels are transmembrane proteins which form membrane pores and open upon specific binding of chemical messengers. We use a planar Patch-Clamp-AFM setup which is a combination of a custom made atomic force microscope, a planar patch-clamp unit (nanJiIon, munich) and an inverse microscope to open ligand gated ion channels with ligands bound to a cantilever. With this setup we immobilize living cells and are able to contact and manipulate them both electrically and mechanically. Therefore we aim to measure simultaneously ion channel currents and binding forces of the ligand to its gating receptor and thus to characterize the channel opening event.

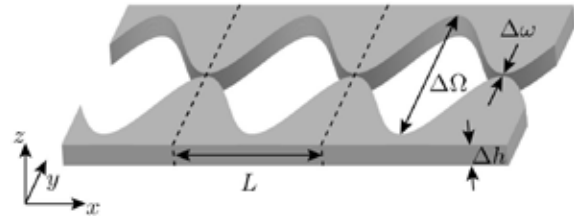
In experiments on the motor protein prestin we already measured the mechanical interaction of cells with the cantilever while applying voltage pulses across the membrane and demonstrated the possibility to measure simultaneously electrical properties of the cell and forces resulting from the electrical pulses. In experiments with functionalized cantilevers we could measure specific interaction between lever bound ligands and binding sites of transmembrane proteins. As ligand gated ion channel we chose the purinergic receptor P2X7 which opens upon ATP binding. Planar patch-clamp experiments with ATP derivatives revealed EDA-ATP as a promising candidate for a channel opening ligand which is covalently linked to the cantilever.

Biased Brownian motion in extremely corrugated channels

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Particle transport in micro- and nanostructured channel structures exhibits peculiar characteristics which differ from other transport phenomena occurring for energetic systems [1]. The theoretical modelling involves Fokker-Planck type dynamics in three dimensions which cannot be solved for arbitrary boundary conditions imposed by the geometrical restrictions. Recently, much effort is drawn on a reduction of the complexity of the problem resulting in the so-called Fick-Jacobs approximation in which (infinitely) fast equilibration in certain spatial directions is assumed. We derive a reduction method [2] which (i) corresponds in zeroth order in the expansion parameter, which



describes the corrugation of the tube wall, to the celebrated Fick-Jacobs result and (ii) extends the validity of the Fick-Jacobs approximation towards extremely corrugated tube structures.

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Artificial Fmoc/Boc- protected amino acids for the solid-phase-assisted synthesis of defined polyaminoamides

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In recent years the design of synthetic carriers for gene delivery has become a research field of high interest. Cationic polymers have shown promising results with polyethylenimine (PEI) being a prominent example, although its toxicity is still an issue of concern. Therefore modified structures have been designed aimed at reducing toxicity and increasing the delivery efficiency. A further disadvantage of PEI is the high degree of polydispersity resulting in a wide range of molecular weight fractions.

Here the construction of Fmoc/Boc- protected artificial polyamino acids which are applicable to standard solid phase synthesis conditions is presented. These new building blocks offer the opportunity to design precise, sequence-defined polyaminoamides. Partial protonation at physiological pH provides positive charges allowing the condensation of negatively charged

nucleic acids like DNA or siRNA. Further protonation at endosomal pH after cellular uptake enables the so-called "proton sponge effect", leading to endosomal release of the carrier into the cytosol.

The synthesis on solid phase provides the advantage of producing defined structures that facilitate the analytic characterization and fulfil an important criterion for a possible clinical application in the future. Based on this approach a library of effective gene delivery vectors was produced in our group. Furthermore, a novel convergent synthesis strategy is presented. This concept should allow to produce polymers of increased molecular weight in higher purity and with less laboratory effort for their use as efficient gene delivery carriers of low toxicity.

Photoluminescence dynamics of quantum dots and quantum wells modulated by surface acoustic waves

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We present a detailed study on the influence of surface acoustic waves (SAW) on the photoluminescence (PL) of self-assembled quantum dots (QDs) and the surrounding 2D wetting layer (WL) [1]. The data is acquired in a conventional micro-PL setup using a stroboscopic excitation scheme of a pulsed picoseconds diode laser which is actively phase-locked to RF signal generator exciting the SAW [2]. We perform numerical calculations which reproduce our experimental findings and provide insights into the dynamics of electrons and holes subjected to the electric fields induced by the high frequency strain fields of the SAW.

The influence of a SAW on the PL of a quantum well has been investigated for more than 10 years. For the WL emission we also observe the characteristic quenching with increasing SAW power, which is explained by exciton dissociation in a Type-II bandedge modulation induced by the SAW. The dissociated charge carrier species are spatially separated at their respective stable points in the bandstructure separated by half the acoustic wavelength and conveyed by the SAW [3,4]. Our phase-resolved technique allows us to resolve the full dynamics of this process

which we study for different SAW power levels: In the low SAW power regime, we observe one PL maximum per SAW cycle which we attribute to the electrons being conveyed, whereas the holes remain at the position at which they are photogenerated. This difference arises due to the different mobilities of electrons and holes. The maximum PL intensity is observed at the local SAW phase at which electrons are generated at a local minimum in the conduction band [4]. In contrast, in the high SAW power regime, we observe two PL maxima per SAW cycle which are separated by 180°. These two maxima are attributed to the stable region for electrons and holes in the conduction and valence band, respectively. These experimental observations are found in good agreement with our numerical modeling of the SAW-induced carrier dynamics.

The emission of individual QDs shows a similar dynamic modulation by the SAW. Different exciton configurations show either one or two maxima per SAW cycle with characteristic relative phase dependencies. These observations can be understood by the dynamic acoustic modulation of the local electron and hole densities at the position of the QD.

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Direct quantification of antibody concentration and affinity in human serum using microscale thermophoresis

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Antibodies are commonly used as indicators for differential diagnosis and therapy monitoring of infections, allergic reactions and autoimmune diseases. Based on Microscale Thermophoresis (MST) we developed an autocompetitive strategy to directly quantify both absolute concentration and binding affinity of antibodies in human serum.

We successfully measured autoantibodies against the cardiac beta-1 adrenergic receptor found in patients suffering from dilated cardiomyopathy. The peptide COR1 was designed to mimic the dominant epitope of the receptor's second extracellular loop. The binding of this fluorescently labeled, artificial antigen to the autoantibodies was analyzed using optically generated thermal gradients. Antibody concentrations from 2 to 200 nM were resolved and the dissociation constant was measured to be 75 nM in 50% untreated human serum.

MST combines the advantages of established binding assays like ELISA (Enzyme-linked Immunosorbent Assay), RIA (Radioimmunoassay) and methods based on FRET (Fluorescence Resonance Energy Transfer). It allows immobilization free, non radioactive measurements in microliter volumes and requires only one fluorescently labeled binding partner. Furthermore, the simple mix and probe protocol minimizes systematic errors. Our autocompetitive approach is not only a promising new tool for serological diagnosis. As it can also be employed for binding analysis in complex biological fluids, for example cell lysate, it is likely to find various applications in the increasingly quantitative field of molecular biology research.

Study the magnetotactic bacteria with optical tweezers

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Magnetotactic bacteria (MTB) are a group of motile bacteria that produce intracellular chains of single-domain nanomagnetic crystals made of magnetite (Fe_3O_4) or greigite (Fe_3S_4). These chains impart a permanent magnetic dipole moment to the cell body, allowing it to navigate along geomagnetic field. Understanding the evolutionary advantage of magnetic navigation and the biomineralization mechanism of single-domain magnetic particles fascinates scientists in biology and material science. MTB also serve as model system for understanding magnetite biomineralization and magnetic navigation in migratory animals such as salmon or songbird. We have developed an "optical tweezers" (OT) system to study the swimming dynamics of MTB. This is a non-invasive technique and can be used in liquid environment. OT use a focused laser beam with Gaussian intensity profile to trap a transparent particle in its focal point and control its position in space. It can apply forces in range

0.1-100 pN and with spatial resolution of 0.1-2 nm [1] which is the range of stoke forces for bacteria. The 1064 nm laser wavelength induces small photo-damage to living cells and allows us to do long term experiments. Our optical tweezers setup is in an inverted configuration, which allows us to combine it with other microscopy and spectroscopy techniques in up-right position; such as optical microscope (current configuration), confocal microscope (for MTB studies), Raman spectroscopy or even AFM. In addition, setup has a Helmholtz-coil system for magnetic manipulation of MTB. Later on, the possibility of trapping a magnetotactic bacterium will be investigated. In all further steps a microfluidic device will be used to increase the accuracy of measurement, control and design the growth condition of bacterium.

[1] Neuman, K. C. & Nagy, A, *Nature Method*, 2008, p.492

Trithiolates on Cu(111) – LEED as a powerful technique for structure determination and phase transition analysis

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Well ordered surface supported organic monolayers are highly topical and many experiments aim at both an atomically precise structure determination and characterization and understanding of frequently observed phase transitions. We demonstrate STM and LEED experiments on aromatic trithiolate molecules on Cu(111) as versatile model systems for both strongly anchored molecules but also metal-coordinated networks based on thiolate-copper-thiolate interconnects.[1] Metal-coordinated networks are prepared from an initial well ordered precursor structure of 1,3,5-tris(4-mercaptophenyl)benzene (TMB) by means of thermal annealing. This initial structure is prepared under UHV conditions by room temperature deposition of TMB onto the reactive Cu(111) surface, whereby all thiol groups become deprotonated and the resulting trithiolate molecules are strongly anchored to the substrate by means of Cu-S bonds. The aim of the present work is obtaining detailed knowl-

edge of this initial structure by quantitative Low Energy Electron Diffraction (LEED) experiments. Structure determination by quantitative LEED experiments includes recording of LEED patterns as a function of electron energy, extracting I(V) curves for several unique reflections from these diffraction patterns, and comparison of experimental I(V) curves to dynamic simulations considering multiple scattering. As in common diffraction experiments, structure refinement is carried out by variation of atomic coordinates in order to obtain the best fit between experimental and theoretical I(V) curves. We also demonstrate that LEED is a powerful technique for monitoring thermally induced adsorbate superstructure phase transitions in real time. In this respect, the chosen system of 1,3,5-benzenetriol (BTT) is particularly promising, because an initial precursor structure undergoes two phase transitions upon thermal annealing.

[1] Walch et al., *J. Am. Chem. Soc.*, 2011, 133, 7909-7915

Functionalization of wire-frame DNA nanostructures

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Nanometer-sized polyhedral wire-frame objects hold a wide range of potential applications both as structural scaffolds as well as a basis for synthetic nanocontainers. The utilization of DNA as basic building blocks for such structures allows the exploitation of bottom-up self-assembly in order to achieve molecular programmability through the pairing of complementary bases. We present here two different areas of work undertaken to utilize such hollow DNA-based frameworks. A structurally variable, multi-functional tetrahedral framework of 75 nm strut length constructed from the DNA origami method. While its rigidity offers use as a precisely measured spacing scaffold, flex-

ible hinges at each of their four joints provide a means for structural variability of the object. The intrinsic site addressability provided by this technique allows the unique targeted attachment of dye and/or linker molecules at any point on the structure's surface, which has made it suitable as a tool for emerging superresolution microscopy techniques such as DNA-PAINT. Additionally, initial work towards the construction of biocompatible DNA-based wire-frame containers is presented. Here, we seek to exploit multi-component self-assembly processes to fuse DNA nanostructures with soft materials and other molecules for the generation of functional constituents.

Development of a Mechanical Single Electron Transistor

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We present progress in the development of a mechanical single electron shuttle. The shuttle is composed of a gold island on a silicon nitride beam situated in a gap between source and drain electrodes. Oscillation of the beam brings the island into contact with the electrodes, and in the presence of a DC

bias charging of the island results in electron transport. The island is equipped with a gate electrode and the motion of the beam can be driven dielectrically. The current design has potential to function as a mechanical single electron transistor at 4K.

Single-molecule four-color FRET visualizes energy-transfer paths on DNA origami

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Directing energy by fluorescence resonance energy transfer (FRET) is utilized by nature in light-harvesting complexes for photosynthesis. To artificially build a system where the path of energy transfer can be manipulated on the nanoscale we used rectangular DNA origami as a molecular breadboard to precisely assemble fluorophores in a desired fashion.[1] In the origami technique one ~7.3 kbases long single-stranded DNA is hybridized with ~200 short synthetic DNA "staple" strands to build a desired structure.[2] By modifying the respective "staple" strands, the fluorophores were incorporated such that the light from the "blue" input dye (ATTO488) could either be guided to the "red" (ATTO647N) or "IR" (Alexa 750) output dye, by a "green" dye (ATTO565) that was placed at two alternative positions.

Specifically, the molecules were sorted with the aid of stoichiometric ratios to select the fully labeled species for further analysis. The calculated FRET-related ratios demonstrate that the position of the green dye determines the energy-transfer path towards the "red" or the "IR" output dye.

To summarize, we show that the energy-transfer path can be manipulated on the nanoscale. The red and IR dyes provide an output signal that can be easily read out. DNA origami might therefore serve as a circuit board for photonic devices beyond the diffraction limit and could additionally offer new possibilities for nanoscale sensors by replacing the jumper strand by sensor strands that could report on binding of biomolecules.

For analysis of the energy transfer paths on the DNA origami, we used a single-molecule four-color FRET approach with alternating laser excitation and individual detection of the four different fluorophores. This setup advances previous multi-color schemes and has the potential to monitor six distances within a biomolecular complex simultaneously, as well as the capability

[1] Stein, I.H., Steinhauer, C. and Tinnefeld, P. *Single-Molecule Four-Color FRET Visualizes Energy-Transfer Paths on DNA Origami*, *JACS*, 133, 4193–4195 (2011). (including cover)

[2] Rothmund, P.W.K. *Folding DNA to create nanoscale shapes and patterns*, *Nature* 440, 297-302 (2006).

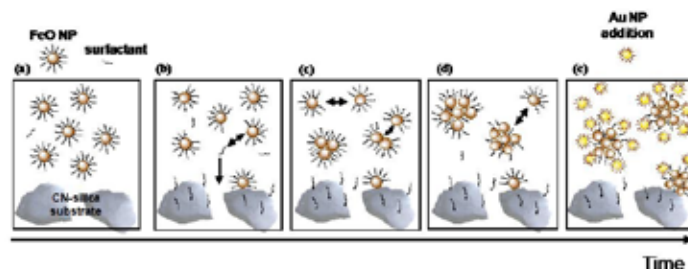
Controlled aggregation of magnetic nanoparticles and formation of gold-iron oxide nanocomposites

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We present a new route to stable magnetic-plasmonic nanocomposite materials with exceptional control over composite size and very high monodispersity. The method involves the assembly of magnetic iron oxide nanoparticles, of any size in the superparamagnetic size range. Their steric repulsion is gradually reduced by competitive stabilizer desorption arising from the presence of a tertiary silica phase. This process is analysed using Smoluchowski aggregation equation and Fuchs stability ratio dependent on nanoparticle-nanoparticle interac-

tions. Subsequent addition of gold nanoparticles results in hierarchical assemblies in the form of gold-decorated magnetic nanoparticle clusters, in a range of possible sizes from 20 to 150 nm, selected by the timing of the addition. This approach potentially adds plasmonic and chemical functionality to the magnetic clusters and improves the physical robustness and processability of the suspensions. Most critically, the clusters remain superparamagnetic in nature. These advantages enhance the potential of the materials as size-selected contrast agents for magnetic resonance imaging.

[1] J. K. Stolarczyk, S. Ghosh, D. F. Brougham "Controlled growth of nanoparticle clusters through competitive stabilizer desorption", *Angew. Chem. Int. Ed.* 2009, 48, 175-178.

[2] C. J. Meledandri, J. K. Stolarczyk, D. F. Brougham "Hierarchical gold-decorated magnetic nanoparticle clusters with controlled size" *ACS Nano* 2011, 5, 1747-1755.

Analytical methods for the physico-chemical characterization of siRNA polyplexes

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Silencing of gene expression by RNA interference offers the possibility to cure diseases that are associated with the over-expression of a gene or the expression of a pathogenic gene. To date, in the field of polymer-based siRNA delivery, the main focus has been on the development of more efficient polymers. For the development of a siRNA-polyplex drug product, it is essential to have reliable characterization techniques to ensure quality, reproducibility and stability of the polyplexes.

Thus, the purpose of this study was to evaluate several analytical methods for the characterization of siRNA polyplexes. Therefore, three sequence-defined polyamidoamines were synthesized through solid-phase assisted synthesis. siRNA polyplexes were prepared by mixing siRNA solution (200µg/ml) with polymer solution (at the ratio of N/P12). After incubation, the formed

polyplexes were characterized using dynamic light scattering (DLS), atomic force microscopy (AFM), fluorescence correlation spectroscopy (FCS) and nanoparticle tracking analysis (NTA). The sizes of polyplexes were 20-40nm (Polyplex 1), around 150nm (Polyplex 2) and 100-430nm (Polyplex 3). Whereas all methods were suitable for medium sized polyplexes of 80-200nm, the size determination of very small polyplexes was not possible with NTA. For large and heterogeneous polyplexes only DLS and AFM led to satisfying results.

In conclusion, the use of comprehensive analytical techniques is required for a complete picture of the siRNA polyplexes, especially in order to characterize samples that contain very small or large polyplexes, as well as mixtures of both.

Photocatalysis with carbon nitride and hybrid materials

Brian Tuffy, Hongji Wang, Prof. Bettina Lotsch

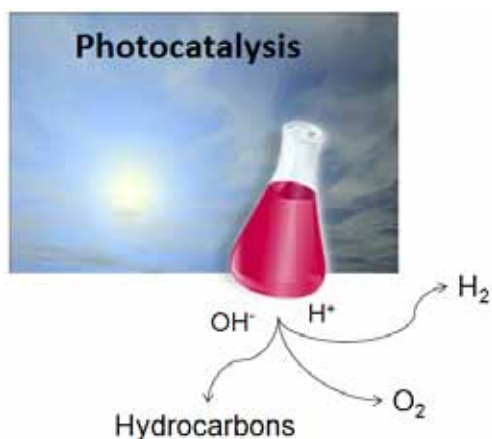
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Carbon nitride (CN_x) and CN-titanate hybrid materials offer great potential for their use in photoreactions such as the reduction of volatile organic compounds, water splitting, CO₂ reduction and efficient photovoltaics. Polymeric carbon nitrides are cheap, functional and easily synthesised materials which have shown promising visible light photoactivity.

In this project, various carbon nitride based materials are synthesised, chemically analysed and their photocatalytic activity measured by gas chromatography. These materials are characterised with FT-IR, XRD, UV-VIS, XPS, NMR and GC. Our carbon nitride materials are also compared to other well known CN_x species. i.e. melon, melon etc.

Interestingly our materials indicate different structures depending on calcination conditions and have distinctly different photocatalytic activity (UV illumination). The best activity is from one of our materials assumed to be previously unreported. It is yet to be seen if this trend changes for visible light illumination and if a correlation exists between the degree of condensation and the bandgap for which photocatalytic activity is observed.

Photoactivity is measured in two separate experiments, water splitting to produce hydrogen and CO₂ reduction resulting in methanol production. The results presented show a detailed study of CN materials and their resulting photoactivity. The effects of synthesis conditions, solution conditions and measurement conditions provide insights into the effect of CN_x doping for new photocatalysts.



[1] Bettina V. Lotsch et al., "Unmasking Melon by a Complementary Approach Employing Electron Diffraction, Solid-State NMR Spectroscopy, and Theoretical Calculations—Structural Characterization of a Carbon Nitride Polymer," *Chemistry - A European Journal* 13, no. 17 (June 2007): 4969-4980

[2] Xinchun Wang et al., "A metal-free polymeric photocatalyst for hydrogen production from water under visible light," *Nature Materials* 8, no. 1 (11, 2008): 76-80.

[3] Kazuhiko Maeda et al., "Photocatalytic Activities of Graphitic Carbon Nitride Powder for Water Reduction and Oxidation under Visible Light," *The Journal of Physical Chemistry C* 113, no. 12 (3, 2009): 4940-4947.

Extracting band-gap from 2-dimensional nanostructures with high spatial resolution

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With advancements in nanotechnology and the ever decreasing size of the structures used in devices, understanding their properties at such diminishing dimensions has gained prime importance. Valence energy electron loss spectroscopy (VEELS) used in scanning transmission electron microscopy (STEM) mode of a transmission electron microscope (TEM) offers an attractive option to ascertain band-gaps locally at high spatial resolution (<2nm). VEELS measurements require a very low full width half maximum of energy for the microscope; hence usage of a monochromat-

ed TEM becomes a necessity. In this work a perovskite structure $\text{KCa}_2\text{Nb}_3\text{O}_{10}$ was investigated for structure and electronic properties variation between bulk and exfoliated forms. Electronic band-gap of the perovskite $\text{KCa}_2\text{Nb}_3\text{O}_{10}$ in bulk and exfoliated forms has been measured. In addition the structure of exfoliated nanosheets has been determined with the help of high resolution transmission electron microscopy (HRTEM) acquired with the help of aberration corrected objective lens system.

Simulation of hydrodynamic interactions in active systems

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The study of systems of active particles aims at revealing generic organizing principles for the emergence and conservation of collective phenomena. Being complex by nature, such research has to some extent to rely on simulations of these systems. These models often suffer from ad-hoc interaction rules for the active particles which are not rigorously justified. Here we propose to use multi-particle collision (MPC) techniques for the study of active systems with hydrodynamic interactions. The MPC tech-

nique is well established for the study of hydrodynamics in the low Reynolds number regime and allows for larger simulation boxes than other techniques. Using the MPC approach for active systems it is also possible to separate the role of hydrodynamics from that of other interaction mechanisms for collective behavior. This method is then applied to study the synchronization of rotating linear polymers.

TEM investigation of material properties of anodized TiO_2 nanotubes

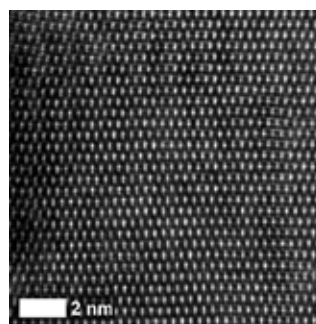
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TiO_2 is a metal oxide with broad application spectrum due to its adaptable electronic and material properties as well as an easy workability. Over the last years, nanostructuring of thin TiO_2 films is an interesting and well-pursued issue, mainly to gain increased surface area and optimized properties, e.g. for implementation in thin-film solar cells.

In this study, we focus on an array of TiO_2 nanotubes which has been fabricated by anodizing a thin film of sputtered Ti on a conducting substrate composed of ITO and a thin TiO_2 compact layer. Transmission electron microscopy (TEM) has been chosen for an in-depth investigation on these nanotubes due to its high resolution and matchless ability in analysis of local material properties. The samples were fabricated using three different anodization voltages to get a basic comparison of the sample characteristics. Bright-field images show increasing dimensions of



the tubes as result of increasing the anodization voltage.

In order to obtain information about the tubes' crystalline features, electron diffraction, dark-field imaging and high-resolution transmission electron microscopy techniques were employed. These methods show considerably larger crystals than expected

from former X-ray diffraction studies, with grain sizes easily exceeding 100 nm in at least one dimension along the tube walls.

These results suggest high electron mobility along the tubes and encourage their implementation in diverse applications.

Exfoliated layered transition metal oxides used as 2D building blocks for new nanostructured hybrid materials

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2D nanosheets such as graphene and exfoliated layered transition metal oxides (TMO) have been emerging recently as new materials with unique structure and size dependent properties.[1-2] With a height of only a few nanometers and a lateral size in the micron range nanosheets show great potential as functional building blocks to construct 3D hybrid superlattices. Tailor-made electronic, optical or magnetic properties can be achieved by the combination of different nanosheets or by variation of their arrangement with nanoscale precision.

Here we focus on the assembly of 'Ca₂Nb₃O₁₀' nanosheets with exfoliated MnII-AlIII-layered double hydroxides.[3] 'Ca₂Nb₃O₁₀'

nanosheets are obtained via common solid-state synthesis of the Dion-Jacobson phase KCa₂Nb₃O₁₀, cation-proton exchange of the as-obtained bulk-material, and exfoliation upon tetrabutylammonium hydroxide combined with mechanical treatment. Flocculated and layer-by-layer arranged samples were characterized by X-ray powder diffraction, scanning-electron microscopy, energy-dispersive X-ray spectroscopy, atomic force microscopy and transmission electron microscopy to demonstrate the wide spectrum of nanomorphologies that can be obtained.

[1] R. Ma, T. Sasaki, *Adv. Mater.* 2010, 22, 5082.

[2] M. A. Bizeto, A. L. Shiguihara, V. R. L. Constantino, *J. Mater. Chem.* 2009, 19, 2512.

[3] S. Werner, B. V. Lotsch, to be submitted.

High-mobility organic thin-film transistors with photolithographically patterned top contacts

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Due to its high resolution, photolithography is the preferred patterning method for pentacene thin-film transistors (TFTs). Since the morphology of thin pentacene films is very sensitive to solvents and heat, the photolithographic patterning of the source/drain contacts is ideally performed prior to the pentacene deposition, which explains the general preference for the bottom-contact TFT structure. However, as experiments and simulations have shown, the bottom-contact structure is associated with larger contact resistance than the top-contact structure, so that top-contact TFTs generally have larger transconductance and higher cutoff frequency than bottom-contact TFTs. The key to realizing high-mobility organic TFTs with photolithographically patterned top contacts is a semiconductor which in the pristine state has transport properties like pentacene, but is less sensitive to solvents and heat than pentacene. An example is dinaphtho-thieno-thiophene (DNNT) which provides mobilities similar to pentacene in shadow-mask-patterned

top-contact TFTs, but unlike pentacene crystallizes in a single polymorph. We have fabricated pentacene and DNNT TFTs with Au top contacts deposited by vacuum evaporation and patterned by either shadow-masking or photolithography. The shadow-mask-patterned TFTs both have mobilities above 1 cm²/Vs (pentacene: 1.5 cm²/Vs, DNNT: 2.2 cm²/Vs). Photolithographic patterning of top contacts on pentacene causes the mobility to drop to 0.16 cm²/Vs, while DNNT is much less affected by photolithography (0.46 cm²/Vs). X-ray diffraction on pentacene and DNNT films before and after photolithography reveal a structural phase change in the pentacene film, associated with a distinct evolution of the pentacene bulk phase at $d = 14.4 \text{ \AA}$. This bulk-phase evolution, presumably initiated by solvent exposure, is correlated with the pronounced drop in mobility in the case of pentacene. The DNNT films, in contrast, are characterized by a single phase, both before and after photolithography.

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OR

Walk to the boat stop "Zattere" and take boat No. 2 (~14 min, every 10 min) or No. 51 (~7 min, every 20 min) to "S. Zaccaria". Then take boat No. 20 to San Servolo.

TIMETABLES

TRAIN TO VENICE AND BACK TO MUNICH

| To Venice (18.09.) | | Back to Munich (23.09.) | |
|---------------------|---------------------|-------------------------|---------------------|
| Munich Main station | Venezia Santa Lucia | Venezia Santa Lucia | Munich Main station |
| 11:31 | 18:10 | 13:34 | 20:25 |

BOAT LINE 20 TO WORKSHOP LOCATION (SAN SERVOLO)

| To San Servolo | | Back to Venice | |
|--------------------|------------|----------------|-------------|
| S. Zaccaria | S. Servolo | S. Servolo | S. Zaccaria |
| 6:55 | 7:05 | 8:35 | 8:45 |
| 7:15 | 7:25 | 8:45 | 8:55 |
| 8:15 | 8:25 | 9:10 | 9:20 |
| 8:35 | 8:45 | 9:40 | 9:50 |
| 9:00 | 9:10 | 10:00 | 10:10 |
| 9:20 | 9:30 | 10:50 | 11:00 |
| 9:50 | 10:00 | 11:20 | 11:30 |
| 10:30 | 10:40 | 12:10 | 12:20 |
| 11:10 | 11:20 | 12:40 | 12:50 |
| 11:50 | 12:00 | 13:30 | 13:40 |
| 12:30 | 12:40 | 14:00 | 14:10 |
| 13:10 | 13:20 | 14:50 | 15:00 |
| 13:50 | 14:00 | 15:30 | 15:40 |
| 14:30 | 14:40 | 16:00 | 16:10 |
| 15:10 | 15:20 | 16:50 | 17:00 |
| 15:50 | 16:00 | 17:30 | 17:40 |
| every 40 min until | | 18:00 | 18:10 |
| | | 18:50 | 19:00 |
| | | 19:20 | 19:30 |
| 20:30 | 20:40 | 20:10 | 20:20 |
| every hour until | | 20:40 | 20:50 |
| | | 23:30 | 23:40 |
| 0:25 | 0:35 | 22:40 | 22:50 |
| 1:30 | 1:40 | 23:40 | 23:50 |

INTERNET

Two PC rooms with internet connection are accessible for the participants located next to the conference hall. Please ask for the keys and login in the conference office next to the lecture hall.

Access to the WLAN internet will be available. Information will be provided at the conference.

Valdese



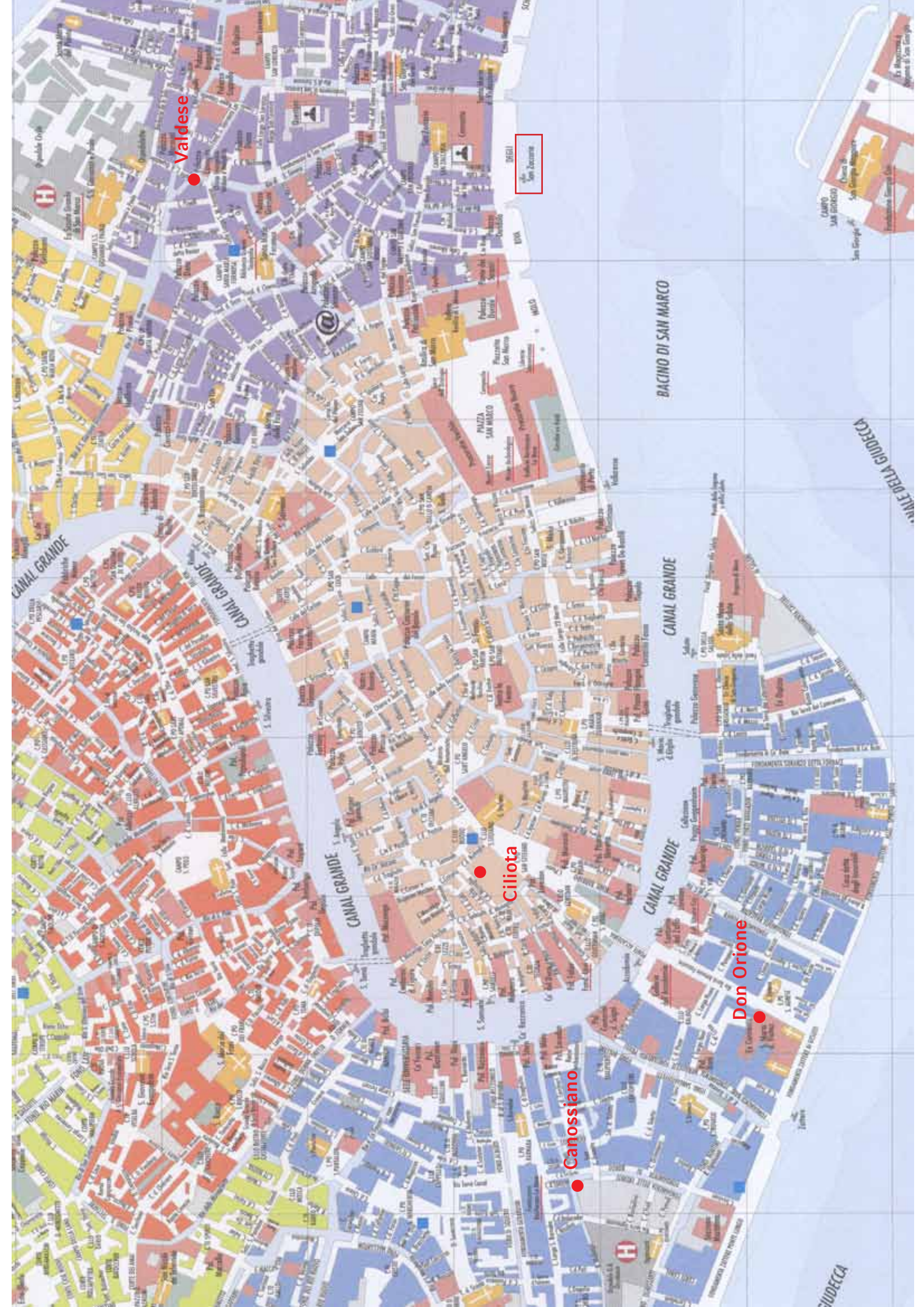
Ciliota



Don Orione



Canossiano



CeNS Workshop 2011: Nanosciences – From molecular systems to functional materials / VIU, San Servolo, (Italy) / September 19 – 23, 2011

| Time | Monday, September 19th Boat departing at 9:00 | Tuesday, September 20th Boat departing at 9:00 | Wednesday, September 21st Boat departing at 9:00 | Thursday, September 22nd Boat departing at 9:00 | Friday, September 23rd Boat departing at 9:00 | Time |
|-------|--|---|---|---|---|-------|
| 09:20 | Daniel Müller Cell mechanics and the biophysics of membrane proteins | Evelyn Hu Gap-mode plasmonic cavities: Engineering light-matter interactions in metallic structures | Paolo Lugli Solution based Carbon Nanotube films for electronics and optoelectronics applications | Klaus Sengstock Quantum simulation of frustrated classical magnetism in triangular optical lattices | Sunny Xie Life at the single molecule level | 09:20 |
| 10:05 | Helmut Grubmüller Conformational dynamics in biological nanomachines: Atomistic simulation and single molecule experiments | Markus Antonietti Artificial photosynthesis and heterogeneous organo-catalysis by carbon nitrides | William Shih Self-assembled DNA-nanostructure tools for molecular biophysics | Bert Nickel An organic dual gate field effect transistor for biosensing | Jörg Schmalian Non-equilibrium dynamics in graphene: Perfect fluid behavior and stimulated emission of photo-excited fermions | 10:05 |
| 10:50 | Coffee break | Coffee break | Coffee break | Coffee break | Workshop ending | 10:50 |
| 11:15 | Gerd Binnig The first moments of STM and some thoughts about the future of nanotechnology | Sebastian Gönnerwein Voltage-controlled spin mechanics | Hiroaki Misawa Plasmon-assisted photocurrent generation system | Stefan Duhr Some like it hot - Biomolecular Interaction Studies using Microscale | | 11:15 |
| 12:00 | Lunch (12:00 h - 14:30 h) | Lunch (12:00 h - 14:30 h) | Allan MacDonald Order in graphene | Lunch (12:15 h - 14:30 h) | | 12:00 |
| 14:30 | Klaus Ensslin Graphene quantum circuits | Carlos Silva Exciton dynamics in semicrystalline polymeric semiconductors | | Uri Banin Hybrid metal-semiconductor nanoparticles; From photocatalysis to doping | | 14:30 |
| 15:15 | Oliver Schmidt Fundamental concepts and potential applications of strainable and shapeable nanomembranes | | | | | 15:15 |
| 16:00 | Coffee break | Posters Session I & Coffee | | Posters Session II & Coffee | Boat leaves at 11:20/12:10 Train to Munich leaves at 13:34 h from train station | 16:00 |
| 16:30 | Hagen Klauk Low-voltage organic transistors and circuits for flexible electronics | | | | | 16:30 |
| 17:15 | Thomas Carell Chemical studies of genome maintenance and stem cell development | Jeff Gore Cooperation, cheating, and collapse in microbial ecosystems | Informal Discussions | Eric Stava Nanomechanical and microfluidic control over ion channels | | 17:15 |
| 18:00 | Welcome reception | Andreas Wallraff Exploring the quantum physics of light with micro- and nanoelectronic circuits | | Jörn Dunkel Hydrodynamics of microorganisms | | 18:00 |