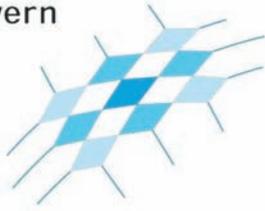


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Macromolecular Systems for Nanoscience – Chemistry, Physics and Engineering Aspects



CeNS-BIMF Workshop 2007

KLOSTER IRSEE, GERMANY, SEPTEMBER 6 – 9, 2007



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INVITED TALKS

Cell-Free Gene Expression on a Chip

Amnon Buxboim, Shirley Daube, and Roy Bar-Ziv

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We are motivated to develop artificial systems based on cell-free gene expression. We have used transcription / translation reactions to develop simple circuit cascades that mimic natural gene networks. We have also shown that protein nano-structures could be synthesized and assembled in such reactions.

A chip with immobilized genes is a natural environment to separate, cascade, and control biosynthetic reactions. A new photolithographic biochip has been developed for the immobilization and expression of genes on a surface and we present a 'structure-function' study of expression on a chip. The study shows how the physical structure of DNA under confinement affects the biochemistry of expression. This synthetic approach may be used to deepen our understanding of heterogeneous biochemical reactions taking place under confinement and crowding in living cells.



Picasso's peace dove imprinted with proteins on a chip.

- [1]. V. Noireaux, R. Bar-Ziv and A. Libchaber, "Principles of cell-free genetic circuits assembly.", *Proc. Natl. Acad. Sci. USA.* 100 (22), 12672 (2003).
- [2]. A. Buxboim, M. Bar-Dagan, V. Frydman, D. Zbaida, M. Morpurgo and R. Bar-Ziv, "A single-step photolithographic interface for cell-free gene expression and active biochips." *Small* 3 (3), 500 (2007).
- [3]. S. Daube, T. Arad and R. Bar-Ziv, "Cell-free co-synthesis of protein nano-assemblies: tubes, rings, and doughnuts.", *Nano Lett.* 7 (3), 638 (2007).
- [4]. A. Buxboim, S. Daube and R. Bar-Ziv, "Gene brushes: control of protein biosynthesis on the nanoscale", submitted.

Tuning Block Copolymer Morphologies with Electric Fields – From Chain Stretching to Order-Order-Transitions

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Electric field-induced orientation of block copolymer microdomains has increasingly become an important tool for generation of highly ordered templates for nanotechnological applications [1].

In this talk, we will show SAXS experiments elucidating the mechanisms and kinetics of block copolymer microdomain alignment.

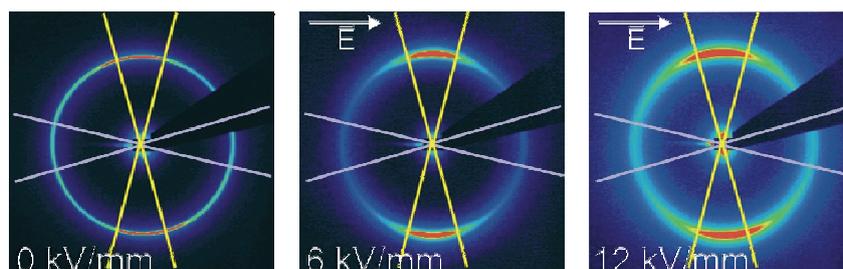


Fig. 1 - Two-dimensional scattering patterns of a 50 wt.% solution of a lamellar PS-b-PI dissolved in THF for different electric field strengths. The arrow indicates the direction of the electric field vector.

In addition, we will demonstrate the distinct influence of electric fields on the microphase behavior, resulting in reversible tuning of the microdomain spacing (Figs. 1 & 2) and even order-order-transitions at sufficiently high field strengths.

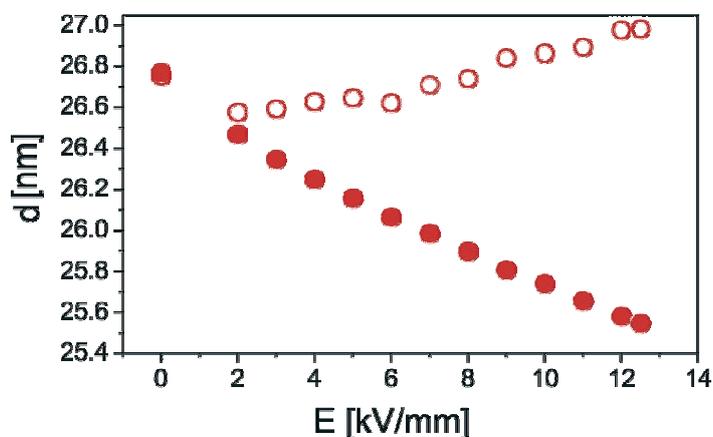


Fig. 2 - Dependence of the lamellar distance d of parallel (●) and perpendicular (○) aligned lamellae with respect to the electric field vector on the electric field strength from the solution in Fig 1.

[1] A. V. Zvelindovsky: "Nanostructured Soft Matter: Experiment, Theory, Simulation and Perspectives" Springer, 199-230 (2007).

Correlation between Chain Morphology and Electronic Structure in Conjugated Polymers

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In the interdisciplinary and vast field of macromolecular science, conjugated polymers are of particular interest, because of their unique electronic structure combined with the mechanical properties of plastic materials. While conjugated polymers are often cited for the semiconducting properties, they constitute equally fascinating systems for polymer physics, because of their fluorescence properties which allow performing optical spectroscopy at the single molecule level.

Among the huge number of currently available conjugated polymers, the attention will be focused on *poly(dioctyl-fluorene)*. This phenylene based polymer represents a model system for structure properties correlations because of its phases (*glassy* and β). These differ for the degree of planarization in adjacent repeat units and show remarkably different luminescent characteristics [1]. Correlations become most apparent at the single molecule level, where the large inhomogeneous broadening of spectral lines can be overcome. By low temperature single molecule polarization anisotropy we have probed the conformation of isolated chains in the *glassy* and the planarized β -phase. The results demonstrate that β -phase chains are characterized by higher polarization values than the *glassy* ones, reflecting a structure comparable to a one-dimensional crystal [2]. Contrary to that, the random values of the dihedral angle between the repeat units in the *glassy* phase lead to a decrease in the overall chain stiffness and consequently reduce the polarization anisotropy. A detailed investigation of β -phase chains shows a strong correlation between the zero phonon

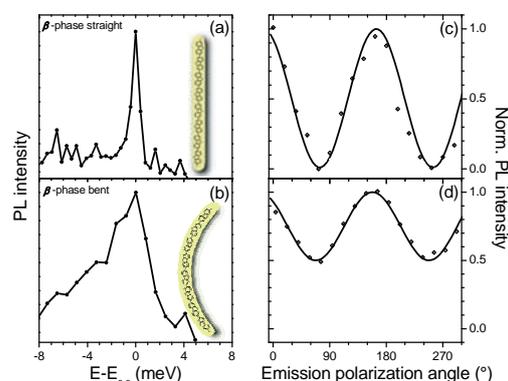


Fig. 1. (a, b) Zero phonon line spectra of two β -phase polyfluorene single chains. (c, d) Corresponding emission polarization anisotropy curves. Insets are the proposed chromophore conformations.

linewidth and the polarization anisotropy in the emission (Fig. 1). Linewidth provides a lower limit for the electronic coherence, a parameter which is crucial for a microscopic understanding of the charge and energy transfer. Polarization anisotropy is symptomatic of the amount of bending in the chromophore. Extremely narrow lines ($< 500 \mu\text{eV}$) are observed for straight chromophores (Fig. 1 a, c). In contrast, larger linewidths are systematically observed for weakly polarized emission (Fig. 1 b, d). The results establish a clear correlation between intrachain morphology and the photophysics. Moreover, the role of electronic coherence in determining the properties of conjugated polymers will be discussed.

[1] K. Becker, J. M. Lupton, *J. Am. Chem. Soc. (Comm.)* 127, 7306 (2005).

[2] E. Da Como, K. Becker, J. Feldmann, and J. M. Lupton, *Nano Lett.* (in press) (2007).

Micromechanics of Ultrathin Polymeric Membranes - From Capsules to Wrinkles

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Mechanical properties of ultrathin polymeric films are of interest not only for stability aspects, but as well for transport properties of nano-objects in flow, adhesion properties or for the design of mechanical switches and valves, to mention just a few examples. Studying these properties requires new experimental approaches. We give two examples of nanomechanical studies on polymeric membranes. In the first case, we report results on measurements for microcapsules fabricated by layer-by-layer deposition of polyelectrolytes and discuss implications for the development of stimuli sensitive microcapsule systems. In the second part, we turn to solid supported membranes and highlight recent developments for controlled wrinkling of these systems. We show that wrinkling can not only be efficiently used to measure elastic moduli, but that it can as well serve as a low-cost alternative to lithographic techniques for producing topographically structured samples.

Slow Transport in Complex Environments

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Spatial heterogeneities often give rise to intriguing slow dynamics in complex materials. Examples constitute colloidal gels with stress-sustaining networks, or the crowded environment of the cytoplasm. A minimal model that encompasses spatial disorder and slow dynamics is provided by the Lorentz model, where a classical point particle explores space in the presence of a frozen array of randomly distributed hard spherical obstacles.

We present extensive molecular dynamics simulations and show that a dynamic scaling hypothesis explains the anomalous relaxation close to the localization threshold [1]. The large crossover regimes are reminiscent to the density-dependent exponents of anomalous transport due to 'molecular crowding' observed in the cell and we suggest that such a behavior is in deed generic.

Furthermore we discuss the long-time tails of the velocity autocorrelation function [2] which have been predicted almost forty years ago, but have eluded computer simulations so far.

The model is extended to a needle in the presence of obstacles in the plane to study the effects of entanglement versus excluded volume. The orientational motion is shown to be consistently described in terms of a tube model in the spirit of Doi and Edwards. In particular, we corroborate the scaling behavior of the orientational diffusion as the entanglement due to the obstacle density increases.

[1] F. Höfling, T. Franosch, and E. Frey, *Phys. Rev. Lett.* 96, 165901 (2006).

[2] F. Höfling and T. Franosch, *Phys. Rev. Lett.* 98, 140601 (2007).

DNA as a Tool for Nanoscience

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With over half a century of intense research into its physical properties and biological functions, DNA has become a remarkably useful material: easy to design, obtain, handle, characterize and even observe at the single molecular level. In this talk I will present a survey of ways in which our group is working with DNA to build devices and probe biological and physical phenomena at the nanoscale. These fall into three general categories: using DNA as a scaffold for assembly of other molecules or functional units; building DNA-based devices to characterize DNA-binding proteins; studying DNA self-assembly in its own right.

Synthesis and Application of Vertically Oriented, Highly Ordered Arrays of TiO₂ Nanotubes

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Highly ordered, vertically oriented TiO₂ nanotube-arrays fabricated by potentiostatic anodization of titanium constitute a material architecture that offers a large internal surface area without a concomitant decrease in geometric and structural order. The precisely oriented nature of the crystalline (after annealing) nanotube arrays makes them excellent electron percolation pathways for vectorial charge transfer between interfaces. In a variety of applications including hydrogen gas sensing, water photoelectrolysis, and in heterojunction solar cells the material architecture has demonstrated remarkable properties worthy of careful consideration. For example, in response to 1000 ppm hydrogen at room temperature the TiO₂ nanotube arrays demonstrate a change in electrical resistance of 8.7 orders of magnitude, that is 50,000,000,000%, the largest known sensitivity of any material, to any gas, at any temperature. Under UV spectrum illumination (320 nm – 400 nm) nanotube arrays \approx 30 μ m long demonstrate an un-optimized light-to-chemical energy photoconversion efficiency of 16.25% [1]. As will be discussed, the anodization chemistry has recently been extended to mixed metal systems, enabling fabrication of n-type Fe-Ti-O and p-type Cu-Ti-O nanotube arrays that strongly absorb in the visible spectrum.

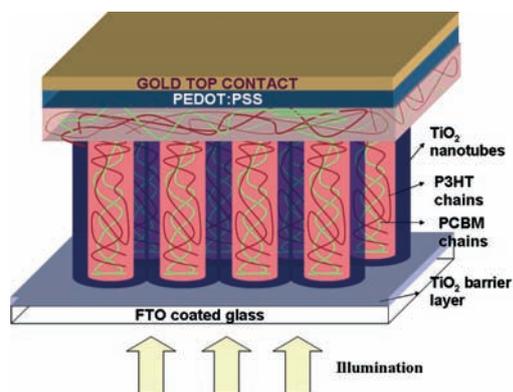


Fig. 1 - Transparent TiO₂ nanotube array based double heterojunction device configuration. Not shown, difficult to draw, is that the polymer percolates in between the nanotubes as well.

As shown in Fig. 1, transparent nanotube array films have recently been used in the fabrication of double heterojunction solid state solar cells [2], with a blend of regioregular poly(3-hexylthiophene) and a methanofullerene (Phenyl C₇₁-butyric acid methyl ester) infiltrated into the transparent nanotube array films. In addition to the P3HT-PCBM interfaces, the walls of the TiO₂ nanotubes provide an additional heterojunction for charge separation. The resulting solid state solar cells show an AM 1.5 short-circuit current density of 12.4 mA/cm², 641 mV open circuit potential and a 0.51 fill factor yielding a power conversion efficiency of 4.1%.

Herein we consider fabrication of the nanotube array architecture, as well as several applications including hydrogen gas sensing, solar generation of hydrogen by water photoelectrolysis, use in solid state heterojunction solar cells, biofiltration, and in enhancing blood clotting to help control hemorrhage.

[1] C. A. Grimes, J. Materials Chemistry 17, 1451 (2007).

[2] G. K. Mor, K. Shankar, M. Paulose, O. K. Varghese, C. A. Grimes. In press, Nature Nanotech.

Controlling Complex Fluids in Microsystems

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Not only the general trend of miniaturizing technical devices, but also the everyday needs of biotechnology and the urge to understand the complex physics of biosystems have driven the development of microdevices for complex matter systems. This talk will present recent achievements in controlling liquids in open and closed microfluidic structures. It will be shown how liquid can be transported in open channels by means of electrowetting. It will be demonstrated that the internal structure of complex fluids may be harnessed by providing suitable prestructures, and how one may exploit these effects to set up combinatorial chemistry labs for picoliter reactant volumes. It will finally be speculated how these systems may be used to design molecular electronic circuitry in a self-assembled way.

Revealing the Interplay of Forces: Dewetting Fluids, Adsorbing Proteins and Sticking Geckos

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Intermolecular forces embrace all forms of matter and are also present in our every-day live as they e.g. determine the strength of a glue or the wetting properties of water on textiles. In my talk I will describe our way to access these forces by studying thin liquid films in the nanometer range [1]. The lessons learned can then be applied to more complex situations like the adsorption of proteins to surfaces like dental implant materials or the adhesion of geckos [2].

[1] R. Seemann, S. Herminghaus, and K. Jacobs, Phys. Rev. Lett. 86, 5534 (2001).

[2] G. Huber, H. Mantz, R. Spolenak, K. Mecke, K. Jacobs, S. N. Gorb, and E. Arzt, Proc. Nat. Acad. Sci. 102, 16293 (2005).

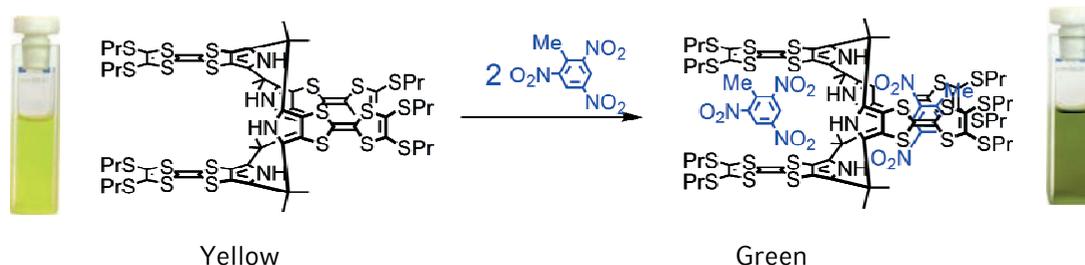
Can an Optical Sensor for TNT Related Explosives be Made?

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² *Department of Chemistry, The University of Texas at Austin, 1 University Station-A5300, Texas 78712 0165, USA*

The advent of supramolecular chemistry [1] has stimulated the contemporary chemists' interest in the development of chemosensors capable of recognizing [2] specific chemical species through weak, non-covalent interactions. The macrocycles, calix[4]pyrroles [3] – first synthesized in the nineteenth century by Baeyer – are endowed with four pyrrole NH hydrogen bond functionalities and have recently received extensively attention as receptors for anionic and neutral substrates. The incorporation of redox-active components into chemosensors is one means of enhancing the recognition process via, e.g., increased donor-acceptor interactions. In this context, the use of tetrathiafulvalene [4] (TTF) appears particularly attractive and a calix[4]pyrrole incorporating four appended TTF units has recently been synthesized [5,6]. It will be demonstrated, that this tetra(TTF)-calix[4]pyrrole is an excellent chemosensor capable of detecting explosives – such as trinitrotoluene (TNT) – commonly used in landmines through a simple color change from yellow to green (see figure below).



- [1] J.-M. Lehn, *Supramolecular Chemistry*, VCH, Weinheim, Germany, 1995.
- [2] (a) P. D. Beer and P. A. Gale, *Angew. Chem. Int. Ed.* 40, 486 (2001). (b) K. A. Nielsen, J. O. Jeppesen, E. Levillain, and J. Becher, *Angew. Chem. Int. Ed.* 42, 187 (2003).
- [3] J. L. Sessler and P. A. Gale in *The Porphyrin Handbook*, Vol. 6 (Eds.: K. M. Kadish, K. M. Smith, R. Guilard), Academic Press, San Diego, 2000, p. 257.
- [4] (a) J. L. Segura and N. Martín, *Angew. Chem. Int. Ed.* 40, 1372 (2001). (b) J. O. Jeppesen and J. Becher, *Eur. J. Org. Chem.*, 3245 (2003).
- [5] K. A. Nielsen, W.-S. Cho, J. O. Jeppesen, V. M. Lynch, J. Becher, and J. L. Sessler, *J. Am. Chem. Soc.* 126, 16296 (2004).
- [6] K. A. Nielsen, W.-S. Cho, J. Lyskawa, E. Levillain, V. M. Lynch, J. L. Sessler, and J. O. Jeppesen, *J. Am. Chem. Soc.* 128, 2444 (2006).

Complementary Coordination of Porphyrin as Scaffold for Biomimetic and Optics Materials

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Imidazolyl-Zn-porphyrin spontaneously dimerizes by complementary coordination with a large stability constant of 10^{11} M^{-1} . The slipped π -stacked dimer structure represents the photosynthetic special pair not only in its close similarity of the structure but also in its photophysical properties. Photoinduced electron transfer produces cation radical delocalized over two porphyrin π -orbitals. Due to the decreased solvent reorganization energy, the charge separation rate is accelerated, while the charge recombination rate is decelerated to promote high charge separation efficiency.

Coupling of two imidazolyl-Zn-porphyrin's at their meso positions leads to one dimensional porphyrin wire of extremely large molecular weights by successive complementary coordination. The excitation energy is not quenched by the wire formation itself and is transferred efficiently through the slipped cofacial dimer by energy hopping even

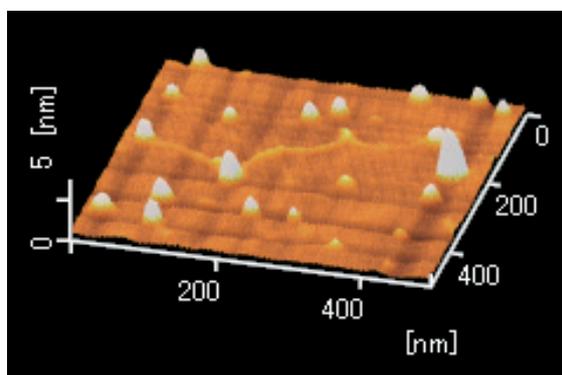


Fig. 1 - AFM image of molecular wire connection of Au nanoparticles.

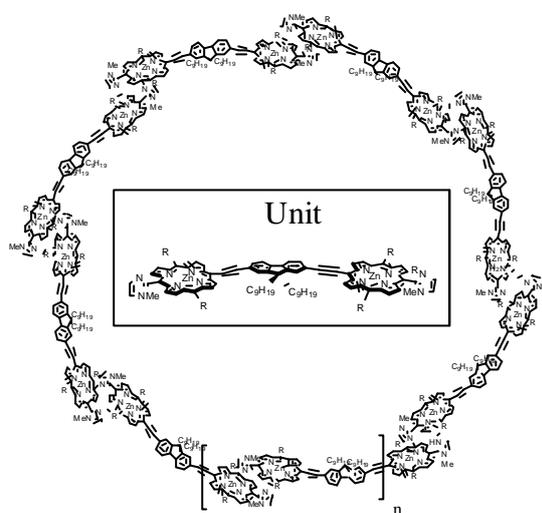


Fig. 2 - Macrocyclization of bisporphyrin bridged by bis(ethynyl)fluorene and its construction unit (inset).

through 50 nm separation to the acceptor attached at the molecular terminals, representing a linear antenna function. Butadiynylene-connected bis(imidazolyl-Zn-porphyrin) was prepared as a molecular wire. This porphyrin array connected gold nanoparticles through complementary coordination to another imidazolyl-Zn-porphyrin attached on the particle via mercaptide modification. The AFM image was shown in Fig. 1.

Connection of two imidazolyl-Zn-porphyrin's by a bent structural unit such as *m*-phenylene leads exclusively macrocyclic structures under dilute conditions. These macrorings are of barrel form. When *m*-bis(ethynyl)phenylene is chosen as the bridging unit, the resulting macrocycle can adopt planar configuration on Au or Cu substrate and clear STM images were obtained. Macrocycle composed of complementarily coordinated dimer unit is an excellent expression of light harvesting antenna complex of purple bacteria. Excitation energy is transferred with 5-20 ps time constants depending on the mutual separation distances by energy hopping process within the ring. The most stable macrocyclic hexamer is accompanied by smaller pentameric macrocycles, since the entropy driven macrocyclization favors the formation of smaller rings. This tendency becomes more evident when the angle is enlarged from 120°. Therefore, diethynylfluorene-bridged bis(imidazolyl-Zn-porphyrin), angle being 160°, gave a maximum distribution at 14- and 15-membered porphyrin macrocycles (Fig. 2) rather than the 18 membered ring of the least angle strain. These large macrocycles are interesting in view of obtaining large two photon absorbing cross sections.

[1] A. Satake and Y. Kobuke, *Tetrahedron*, 61, 13 (2005).

[2] K. Ogawa and Y. Kobuke, *J. Photochem. Photobiol. C*, 7, 1 (2006)

Design of Polymer-Coated Nanoparticles for Positioning in Block Copolymers

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End-functional polymers grafted to form brushes on inorganic nanoparticles can be designed to provide useful fundamental test systems for block copolymer/nanoparticle self-assembly. Short thiol terminated polystyrene (PS-SH), poly(2-vinylpyridine) (P2VP-SH) and PS-*r*-P2VP-SH ($M \sim 3$ kg/mol) were synthesized to allow Au nanoparticles (~ 1.5 nm radius Au core) coated with polymer brushes to be prepared. The polymer brush coating enabled these nanoparticles to be incorporated at low volume fractions into PS-*b*-P2VP diblock copolymers with precise control over nanoparticle location, namely wholly within the PS or P2VP domains or segregated either strongly or weakly to the PS-*b*-P2VP interface. Such control is achieved with three different types of brushes on the Au nanoparticles: a.) High areal chain density (> 1.5 chains/nm²) mixed brushes of PS-SH and P2VP-SH with fractions f_{PS-SH} of PS-SH ranging from 0 to 1; b.) High areal chain density brushes of PS_{*x*}-*r*-P2VP_(1-*x*)-SH random copolymer with different *x* values; c.) Variable areal chain density brushes of PS-SH. For mixed brushes we find that the nanoparticles are segregated to the interface over a wide range (0.90 to 0.10) in f_{PS-SH} while for the PS_{*x*}-*r*-P2VP_(1-*x*)-SH random copolymer brushes *x* must be close to 0.5 before these segregate to the block copolymer interface. These results strongly suggest that the PS-SH and P2VP-SH ligands are mobile on the nanoparticle surface and phase separate in the 2D brush to form "Janus" particles that bind more strongly to the interface than those coated with random copolymer brushes. For the variable areal chain density brushes of PS-SH we find that at areal chain density above a certain value that increases with decreasing molecular weight *M* of the brush, the nanoparticles are segregated to the center of the PS domains while below this value they are segregated to the interface. The preferential interaction between the P2VP block of the PS-*b*-P2VP and the imperfectly shielded Au surface is responsible for the segregation of the nanoparticles with a low areal chain density brush to the interface. The transition areal chain density is $\sim 4/R_g^2$ for $M \sim 3$ kg/mol PS-SH where R_g is the radius of gyration of chains of $M = 3.4$ kg/mol but unexpectedly scales as $M^{-0.55}$ when *M* is varied. Along with suggesting an explanation for this scaling law, I will discuss the changes in block copolymer morphology that occur as the nanoparticle volume fraction ϕ is increased for nanoparticles that segregate to the domain center as well as those that segregate to the interface, the latter behaving as nanoparticle surfactants.

Wormlike and Glassy Wormlike Chains

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The *wormlike chain* is the standard model of a semiflexible thread-like macromolecule. As such it is the basis for our mathematical understanding of the universal mechanical and dynamical properties of important biopolymers such as DNA, F-actin, microtubules, and their solutions and networks. In its scaling limit, known as the *weakly-bending rod*, it admits static and dynamic scaling solutions much like the more familiar Gaussian flexible polymer models.

After a general introduction, the lecture will highlight two recent theoretical developments: the first motivated by – and concerned with – the non-linear non-equilibrium dynamics of single DNA molecules subject to strong external fields [1]; the second, called the *glassy wormlike chain* [2], accounts for the notoriously complicated intermolecular interactions in a semi-dilute solution of (sticky) polymers, such as the cytoskeleton, on a semi-phenomenological level. (As an example for an application, Fig. 1 displays the dynamic structure factor of actin solutions, fitted by the model [3].) Important implications, such as “rheological redundancy” and a generalized time-temperature superposition principle, for the soft glassy rheology of cells and polymer solutions and networks are outlined.

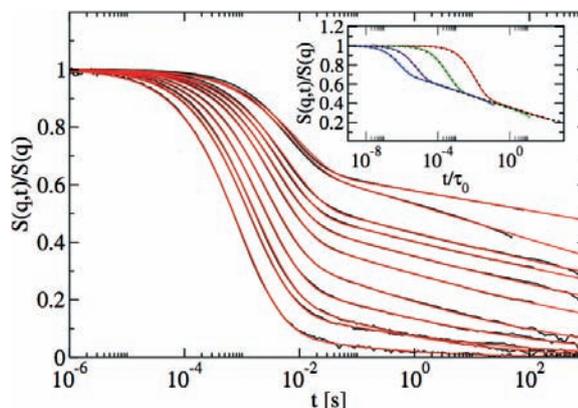


Fig. 1 Dynamic structure factor for pure F-actin solutions at $T = 15^\circ\text{C}$ for various scattering vectors q and the predictions of the GWLC (red lines) [3]. Inset: By exploiting the superposition principle, data for different concentrations can be combined into a master curve extending over up to 10 orders of magnitude in time.

[1] O. Hallatschek, E. Frey, K. Kroy Phys. Rev. Lett. **94**, 077804 (2005).

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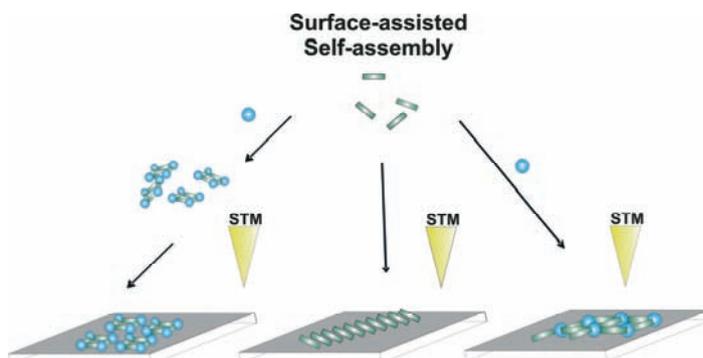
[3] C. Semmrich, J. Glaser, T. Storz, R. Merkel, K. Kroy, A. Bausch, under review at the Proc. Nat. Acad. Sci.

Surface-Confined Self-Assembly

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Molecular nanostructures have recently attracted interest in view of their potential to host tuneable functionalities (e.g. electronic, magnetic, optic etc.), which might be accessible within the few nanometre regime. On the controlled generation of molecular nanostructures on surfaces by coordinative bond formation using open spin metal ions and appropriated organic ligands will be reported. The obtained molecular networks will be investigated at the single-molecule level by means of UHV-STM-techniques carried out at low temperatures.



A combined self-assembly/deposition approach yields a new protocol for the generation of modular metal-ion arrays on surfaces and advents features of a novel, "surface-assisted" coordination chemistry involving magnetically active molecular bricks. Alternatively, the deposition of conventionally synthesized magnetic molecules onto electrically conducting, metallic surfaces will be shown. From this starting point, first experiments directed towards the addressing magnetism at the "single molecule" or "single ion" level will be discussed. The controlled design of operable magnetic surfaces by the principles of surface-assisted coordination chemistry will be a fascinating combination of new scientific perspectives with the aesthetic beauty of the real time observation of the altered interfaces.

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[3] M. Ruben et.al., *Angew. Chem. Int. Ed.* 44, 7896 (2005).

[2] N. Lin, et.al., *Dalton Trans.* 267-280 (2006).

[3] M. Ruben et. al., *J. Am Chem. Soc.* 128, 15644 (2006).

[4] S. Stepanow et. al., *Angew. Chem. Int. Ed.* 46, 710 (2007).

Tailor-Made Surfaces for Microsystems and Nanotechnology

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As devices are getting smaller and smaller and are now approaching even molecular dimensions the influence of the surfaces on the overall performance of a system is becoming more and more important if not dominant. On the other hand, only in a few cases materials can be found that fulfill the requirements needed for the construction of a device and that also provide the necessary surface properties and coatings are needed to optimize the surface properties (Figure 1).

In this contribution we report on chemical strategies that are based on surface-attached polymer layers that allow for the tailoring of surface properties and that also exhibit outstanding stability even in adverse chemical environments. A number of synthetic strategies for the preparation of such layers will be discussed in terms of the specific features and film thicknesses of the resulting layers as well as on ways how to generate microstructures or how to deposit such layers on surfaces within a device that are not well accessible for standard techniques of layer preparation.

Furthermore, systems will be reported in which stimulus responsive polymer carpets are used for the manipulation and movement of nanoparticles on surfaces. Finally, we will also show how the integration of nanometer scale coatings with nanometer rough surfaces leads to interesting wetting properties.

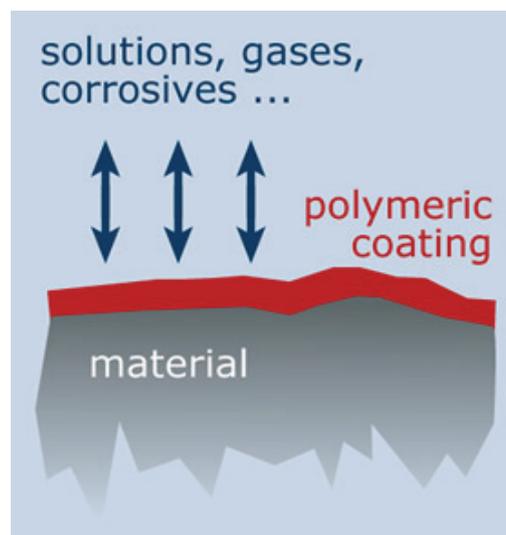


Fig. 1 Materials interact with their environment through surfaces. Thin polymeric coatings can be used to control these interactions.

Nondisruptive Block Copolymer-Based Nanolithographic Processes

T. P. Russell

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As the size scale of features continue to shrink in devices, the use of self-assembly, i.e. a "bottom up" approach, for device fabrication becomes increasingly important. Yet, simple self-assembly alone will not be sufficient to meet the increasing demands placed on the registry of structures, particularly nanostructured materials. Several criteria are key in the rapid advancement and technology transfer for self-assembling systems. Specifically, the assembly processes must be compatible with current "top down" approaches, where standard photolithographic processes are used for device fabrication. Secondly, simple routes must be available to induce long-range order, in either two or three dimensions, in a rapid, robust and reliable manner. Thirdly, the in-plane orientation and, therefore, ordering of the structures, must be

susceptible to a biasing by an external, macroscopic means in at least one, if not two directions, so that individual elements can be accessed in a reliable manner. Block copolymers, specifically block copolymers having a cylindrical microdomain morphology, are one such material that satisfy many, if not all, of the criteria that will be necessary for device fabrication. Here, we discuss several routes by which these versatile materials can be used to produce arrays of nanoscopic elements that have high aspect ratios (ideal for templating and scaffolding), that exhibit long-range order, that give access to multiple length scale structuring, and that are amenable to being biased by macroscopic features placed on a surface.

Block Copolymer Blends for Holographic Data Storage

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Holographic data storage is the most promising technology in the field of optical data storage. In contrast to conventional CDs or DVDs the entire volume of the medium is utilized instead of a single layer. Thus a data storage capacity of several terabytes on a CD-sized disk is proposed. Write-once media, which are mainly based on photopolymer systems, seem to be fairly advanced [1]. Azobenzene-based polymer systems represent a class of a rewriteable media. This presentation reflects the development from basic material considerations of azobenzene-based polymer systems to the processing of the storage media and the photophysical processes to create and store a huge amount of holographic gratings on the same spot by angular multiplexing (AM).

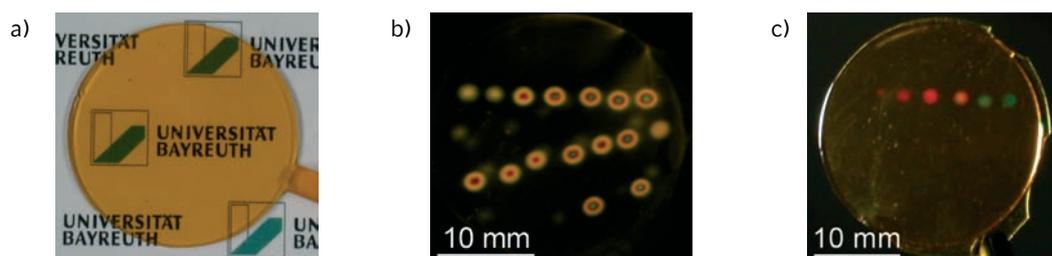


Fig. 1 - a) Picture of an injection-molded polystyrene-based block copolymer blend sample with a diameter of 25 mm and a thickness of 1.1 mm. b) Sample with several inscribed holograms between crossed polarizers. c) Sample showing the first diffraction order of some of these holograms under illumination with white light

Azobenzene-based polymers undergo a photoinduced orientation via multiple trans-cis-trans cycles during illumination by polarized light. Homopolymers or copolymers additionally tend to form surface relief gratings (SRGs), which are detrimental for holographic volume gratings. Such SRGs can be efficiently suppressed by utilizing block copolymers, where mass transport through a rigid matrix is precluded, but the confinement is maintained. This confinement of the azobenzene moieties is essential for the stabilization of the holographic gratings. In polymers based on amorphous azo-moieties, this stabilization is mainly dominated by the T_g of the photoaddressable segment [2]. For AM, thick films in the millimeter range are required, while the optical density (OD) at the writing wavelength has to be in the range of 0.7. By blending such block copolymers with a majority-based, non-absorbing homopolymer dilution of the chromophore minority segment can be achieved. The spheres are randomly distributed, but the sphere size is maintained [3] as well as the photophysical behavior. Samples of 1.1 mm thickness with an adjusted OD can be prepared by injection molding (Fig. 1). Superimposing of several 100 holographic gratings at the same spot was demonstrated as well as 1000 writing and erasing cycles [4]. The long term stability of the stored information exceeds more than two years at ambient temperature [5].

This work was supported by the German Science Foundation in the framework of SFB 481 (projects B2 and Z4).

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Nanostructures for Photovoltaics

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Examples for nanostructures for photovoltaics are mesoporous TiO₂ films in dye-sensitized solar cells. In this presentation recent developments in solid-state dye-sensitized solar cells will be discussed, including investigations of the interfacial engineering on the TiO₂/dye/hole-transporter interface and the influence of the chemical structure of the dye on the solar cell performance.

Furthermore different metal oxide nanostructures will be presented and their possible applications discussed. We propose the use of these structures as electrode materials in hybrid solar cells. Their possible advantages over conventional mesoporous layers will be discussed. Another application of nanostructures for biological and chemical sensors will be briefly presented.

DNA-Switchable Supramolecular Structures

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Supramolecular structures composed of DNA can be switched between different conformations or states by the addition or removal of nucleic acid effector strands. Three different examples for such switchable systems will be discussed: i) switchable DNA aptamers which can controllably bind or release proteins; ii) DNA-crosslinked gels which can be used to reversibly trap and release nanoparticles; and iii) DNA nanomechanical devices which are driven by RNA effector molecules.

Multifunctional Donor-Acceptor Block Copolymers for Solar Cell Applications

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The field of organic and polymer technology for electronics, solar cells etc are gaining more and more importance due to the inherent advantages of cheap production processes and non-conventional design of device structures. The complete control of versatile functions such as light absorption, exciton diffusion, charge separation, charge transport and charge collection at the respective electrodes in an organic solar cell can be achieved by designing multifunctional block copolymers which build up nanostructured bulk heterojunction between donor and acceptor segments via self-assembly. This concept is here presented with suitable examples in which the importance of control of molecular structure up to control of self-assembly and control of device functions are demonstrated. In certain examples the importance of achieving the domain size suitable to the exciton diffusion length will be shown to give efficient photovoltaic devices.

List of poster presentations (short introductions on Friday, 15:15, each 3 min)

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- 2. Bernet, Andreas**
Amphiphilic Low-Molecular-Weight Organogelators and Their Application in Dye-Sensitized Solar Cells
- 3. Friedrichs, Eike**
Control of Genetic Expression on the Transcriptional Level
- 4. Heinz, Paul & Mögele, Florian**
Nanopatterned Surfaces by Molecular Self Assembly - a STM Study
- 5. Hildner, Richard**
Revealing the Electron-Phonon Coupling in a Ladder-Type p-Conjugated Polymer by Single-Molecule Spectroscopy
- 6. Hofmann, Christiane**
Energy and Electron Transfer in Organic Donor-Acceptor Dyads Containing Perylenebisimide Studied by Time-Resolved Spectroscopy
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- 11. Lohwasser, Ruth**
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- 12. Mayilo, Sergiy**
Nanocrystal-Encoded Microbeads for Diagnostics of Autoimmune Diseases
- 13. O'Neill, Patrick**
Sequence-dependent Fluorescence of DNA-hosted Ag Nanoclusters
- 14. Salis-Soglio, Cosima von**
Electret Additives for Isotactic Polypropylene
- 15. Scheler, Esther**
Fluorene Based Reactive Mesogens for Photopatterning
- 16. Schlüsche, Peter**
The mechanism of Gene Regulation Revealed by SpFRET
- 17. Schneider, Christian**
Coagulation Kinetics of Spherical Polyelectrolyte Brushes in Presence of Multivalent Counterions
- 18. Schöffel, Markus**
Imaging SNARE Mediated Membrane Fusion Events with Single Vesicle Resolution
- 19. Schwaderer, Peter**
Pulling on Siloxane Molecules
- 20. Shankar, Karthik**
Hydrogen Production by the Photo-cleavage of Water using TiO₂ Nanotube Arrays
- 21. Sobey, Tom**
From DNA-based Nanotubes to DNA-wrapped Carbon Nanotubes
- 22. Walther, Andreas**
Compartmentalized Cylindrical Nanoparticles – Preparation, Solution Properties and Side-Specific Decoration with Inorganic Nanoparticles
- 23. Yurtsever, Ayhan**
Frequency Modulation Torsional Resonance Mode AFM (FM-TR-AFM) for the In-plane Surface Properties Measurement

POSTER ABSTRACTS

Self-Assembled Nanostructures as Nucleating Agents for Polyvinylidene Fluoride

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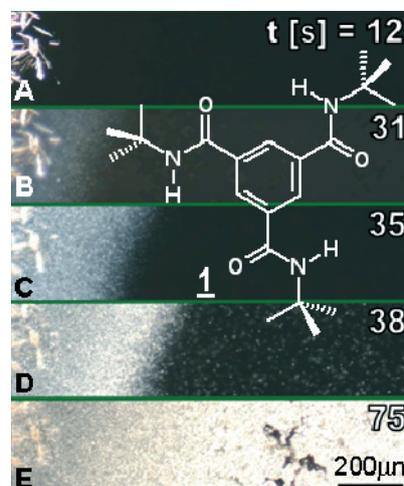
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We present the investigation of substituted 1,3,5-benzenetrisamides as nucleating agents for PVDF. Selected derivatives of this class of compounds are known to be very efficient nucleating agents and clarifiers for isotactic polypropylene [1]. Among in the literature reported nucleating agents for PVDF are PTFE particles and flavanthrone, which is of yellow color [2].

In order to screen potential candidates diffusion experiments were carried out. In the right figure sections of polarized optical micrographs at different times during the isothermal crystallization at 138°C are shown. The samples were prepared by placing tiny amounts of additive whiskers in the middle of a thin (~100 nm) compression molded PVDF film on a microscope slide. This sample was covered, held at 280°C for 10 minutes and cooled to 138°C.

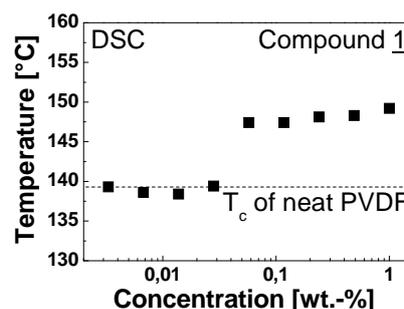
The sections show:

- A: nucleating agent (left) in the polymer melt;
- B: self-assembly of additives;
- C: PVDF crystallization on the additive surface;
- D: PVDF crystallization in the area without additive;
- E: the finally crystallized sample.



Promising compounds have been selected and investigated in mixtures with PVDF to study the concentration dependence nucleating properties.

The diagram on the right shows the crystallization temperature (!) of PVDF mixtures comprising amounts between 1 wt% and 70 ppm of compound 1. Temperatures for crystallization were determined at the minimum of second cooling (10 K/min) after 5 min isothermal heating at 250°C. The crystallization temperatures of the mixtures comprising compound 1 is increased about 10 °C for 1 wt% and still 8 °C for 570 ppm. At lower concentrations no nucleation of PVDF is observed.



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Amphiphilic Low-Molecular-Weight Organogelators and Their Application in Dye-Sensitized Solar Cells

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Low-molecular-weight organogelators are organic molecules capable of growing from homogeneous solution into fine fibrillar structures within an organic solvent. The resulting three-dimensional network yields a macroscopic gel. This self-assembly process is driven by specific non-covalent intermolecular interactions, and the formed physical gels are therefore thermoreversible. Intensive effort has been expended to understand the structure-property relations in order to tailor organogelators; however, up to now it has not been possible to predict in advance if a new molecule will gel an organic solvent or not. Therefore, in recent years, many researchers have attempted to find new efficient gelators, resulting a variety of classes of small molecules with gelling potential.

Conceptually, our amphiphilic low-molecular-weight gelators consist of an apolar cyclohexyl head group, a hydrogen-bond-forming segment based on p-phenylenediamines with amide and urea groups, and an alkyl chain [1]. The gelation behavior of these asymmetrically substituted p-phenylenediamides depends on a well-balanced relation of the terminal head group, the units involved in hydrogen bonding, and on the length of the alkyl chain. With this class of gelators it is possible to tailor thermal and mechanical properties in different organic solvents for various utilizations [2]. This new class of low-molecular-weight amphiphilic compounds is able to gel organic solvents over a broad range of polarity. Especially polar solvents such as valeronitrile, γ -butyrolactone and ionic liquids can be gelled even at concentrations far below 1 wt%.

A promising application is the gelation of ionic liquids in dye-sensitized solar cells (DSCs) (See Fig. 1). DSCs with ionic liquids face the problem of long-term stability due to corrosion and leakage. Simple increase of the viscosity of the ionic liquid leads to reduced performance. Organogelators offer the possibility to yield a quasi-solid state DSC with enhanced long-term stability and maintained performance. We were able to use one of our gelators to prepare a DSC with high efficiency (6.3 % at full sunlight irradiation) and long-term stability (light soaking accelerating stress test at 60 °C over 1000 h) without loss of performance [3].

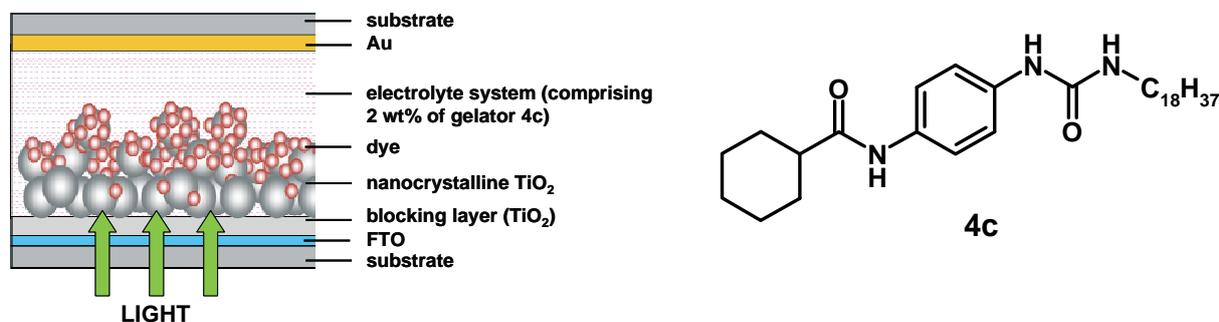


Fig. 1 - Schematic representation of a quasi-solid-state DSC.

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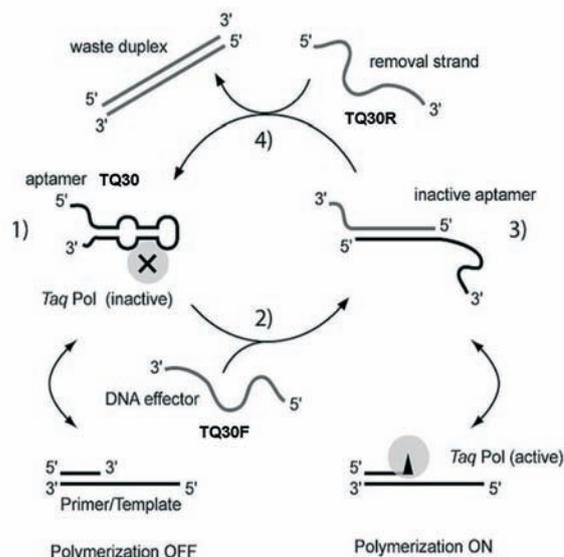
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Control of Genetic Expression on the Transcriptional Level

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Genetic expression comprises the process of turning genetic information - as it is stored in DNA - into biologically functionable proteins. Regulation of expression occurs at any stage of this process. Yet, regulating expression in synthetic systems - even at the most basic level, the level of transcription - remains a challenge. We aim to control the synthesis of mRNA by interference of the transcriptional process by functional DNA or RNA sequences that, for instance, have the capability of binding to and thereby inhibiting polymerases or shield promoter sequences of genes. We demonstrate how control of the enzymatic activity of Taq polymerase is gained by switching a DNA aptamer from an enzyme binding and inhibiting conformation to a non-inhibiting duplex conformation and vice versa [1].



We thank the Deutsche Forschungsgemeinschaft (Emmy Noether grant SI 761/2-3), Nanosystems Initiative Munich (NIM) and the Human Frontier Science Program (HFSP) for their financial support.

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Nanopatterned Surfaces by Molecular Self Assembly - a STM Study

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In the last decades top down engineering via lithographic methods led to high performance functional devices. But it is predicted that the now used common methods will reach their limits at structures below 20 nm. Contrary self assembly is a powerful method to obtain even smaller defined functional structures and has found use in miscellaneous fields such as chemical sensors [1], biotechnology [2], or catalysis and electrochemistry [3]. Self assembled monolayers of metal organic compounds - especially imine bridged complexes - are very interesting because of their catalytical - and in the case of compounds bearing two or more metals - of their paramagnetic properties.

Here we describe the synthesis of a self organizing ligand systems based on 2,4-dihydroxybenzaldehyde [4] and 2,4-dihydroxyacetophenon as well as the corresponding mono- and binuclear complexes. Furthermore derivatives of gallic acid bearing long alkyl chains and UV sensitive units were prepared. The monolayers of the named compounds were investigated at the liquid/HOPG interface using scanning tunneling microscopy. The design of the ligand systems led to highly ordered lamellar 2D structures which are dependent on the alkyl chain length and on the metals contained. The assembly of monolayers is driven by attractive van der Waals interactions of the alkyl chains and occurring H-bonds between

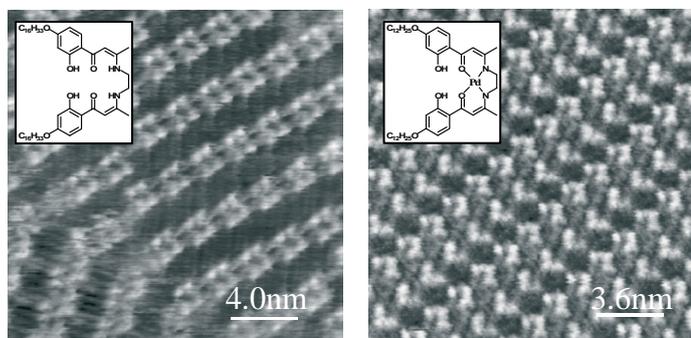


FIG. 1 STM image of an Extended Salen ligand and its corresponding Pd-complex

neighbouring molecules. Thereby the array of the pure ligands can easily be distinguished from the pattern of the corresponding metal complexes.

We are deeply grateful for the financial support by the SFB 569 of the Deutsche Forschungsgemeinschaft (DFG).

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Revealing the Electron-Phonon Coupling in a Ladder-Type π -Conjugated Polymer by Single-Molecule Spectroscopy

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The efficiency of charge- and energy-transfer processes in π -conjugated polymers is strongly determined by their electron-phonon coupling strength. However, the inherent pronounced structural disorder as well as the strong spectral diffusion in this class of functional materials prevented a direct determination of this parameter as yet. Here we employ single-molecule spectroscopy in combination with pattern recognition techniques which allows us to retrieve the profile of the electronic spectrum and concomitantly the electron-phonon coupling strength in a methyl-substituted ladder-type poly(para-phenylene) (MeLPPP). From these data we find a weak electron-phonon coupling for this polymer at low temperatures consistent with the fast excitation energy transfer processes that have been observed for this polymer. Moreover, we find strong evidence that the low-frequency vibrational modes, which are coupled to the electronic transitions, stem from vibrations of the surrounding host matrix indicating an even weaker intra-chain electron-phonon coupling in MeLPPP.

Energy and Electron Transfer in Organic Donor-Acceptor Dyads Containing Perylenebisimide Studied by Time-Resolved Spectroscopy

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We investigated energy and electron transfer processes in organic donor-acceptor dyads by time-resolved fluorescence spectroscopy. Fluorescence lifetimes were determined using a frequency-doubled pulsed Ti:Sa laser system for excitation and a streak camera system for detection, allowing a temporal resolution of up to 2-3 ps. All lifetimes were measured in dilute solution.

In the first type of dyad, perylenebisimide (PBI) is covalently linked to a tetraphenylbenzidine (TPD) unit by an alkyl spacer. TPD acts as an energy transfer donor, PBI as an acceptor. Fluorescence lifetimes of both TPD and PBI have been determined for three different types of spacer and as a function of the solvent. We found that changing the spacer length had a significant influence on

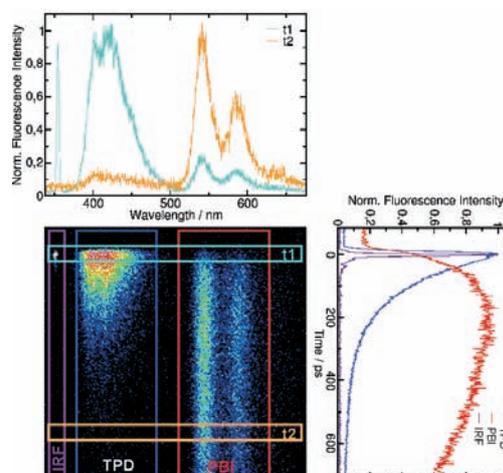


Fig.1: Bottom left: Streak camera image of a TPD-PBI dyad (IRF = instrument response function) Bottom right: Fluorescence decay curves of TPD and PBI. Top: Fluorescence spectra of the dyad at times t1 and t2

the TPD lifetime and on energy transfer efficiency. The PBI lifetimes in all three TPD-PBI dyads were considerably shorter than those measured for pure PBI [1] and varied significantly upon changing the chemical composition of the spacer. We could also influence the PBI lifetimes noticeably by using different solvents. Specific solvent interactions seem to play an important role here, since the lifetime variations could not be attributed to changes in solvent polarity only. The effects described above seem to hint at an electron transfer process between TPD and PBI.

In a second type of dyad, PBI units are covalently linked to fullerenes. Here, PBI acts as an energy transfer donor. The PBI fluorescence lifetime is very strongly quenched in this kind of dyad due to energy and electron transfer to the fullerene.

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Optothermal DNA-Melting in Gold Nanoparticles Aggregates

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Abstract not available.

Towards a Biological Sensor

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In order to measure electronic properties of biological systems, the sensing area has to be adjacent to a sample surface to enable a proper coupling. Here, we investigate the possibility to use organic field effect transistors as a biological sensor. Currently the most controlled way to fabricate such devices is molecular beam deposition of crystalline thin films of Pentacene. The first major obstacle is the operation of a Pentacene thin film transistor (TFT) under water. Therefore the device has to be sealed with an ultra thin capping layer without losing its functionality. First capping experiments and device characteristics are presented. This work is part of an EU-initiative (Biodot).

All-dry Negative Tone Photoresists based on Coumarin Derivatives

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In our days photoresists are used for fabrication of nanoscale patterns. They are usually prepared by spincoating a film from solution and after several steps including irradiation a pattern is realized by using an aqueous or organic developer [1]. An unusual way for preparation of thin photopolymerizable films is physical vapor deposition. In comparison to spincoating this method has the advantage of less impurities and the ability to create uniform films in nanometer thickness range [2]. In addition with this method poorly soluble substances are applicable for thin film preparation. Furthermore, combinatorial approaches can be easily applied [3]. While developing the normal liquid developers can have an impact on the structures due to percolation. To avoid this development can take advantage of the evaporation temperature increase due to the dimerization or polymerization of monomers. Therefore the residual monomer is removed via heating under high vacuum.

In this poster we present an all-dry approach for negative tone photoresists. Since we are using for the first time synthesized coumarin derivatives, there is no photoinitiator necessary. When exposed to UV light (300 – 400 nm), the coumarin group forms, even in the solid state, a cyclobutane ring with a second coumarin group and thus a dimer (see Fig. 1).

In UV spectra, this reaction can be easily followed by the two decreasing and finally vanishing characteristic peaks at wavelengths of 285 nm and 314 nm caused by the disappearance of the double bond.

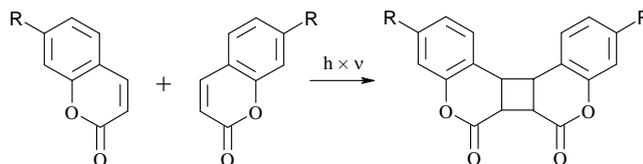


Fig. 1 Dimerization of two coumarin groups by photocyclization [4].

We synthesized mono-, bi- and trifunctional coumarin derivatives for investigations. These were evaporated onto a primed silica wafer, patterned using a mask and UV light, and developed by heating under high vacuum to evaporate the unexposed non-polymerized molecule. The coumarin derivatives need for patterning in solid state a very high exposure time. In addition the developed features showed residues of, in the development step recrystallized, monomer.

To solve these issues, we synthesized a sterical hindered monofunctional coumarin derivate **1**. This was coevaporated with the sensitizer Michler's ethyl keton **2**. Due to this the obtained films needed a significant lowered exposure time.

Fig. 2 shows that the coumarin derivate **1** is vapor depositable and patternable without the use of any solvents. Thus this method has a high potential for all-dry negative photoresist systems.

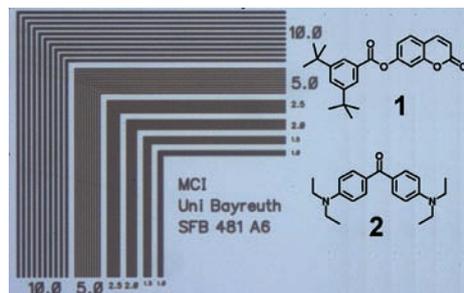


Fig. 2 Reflection microscope image of a developed coumarin/sensitizer resin.

Acknowledgement: We wish to thank the Deutsche Forschungsgemeinschaft, SFB 481, project A6 for financial support of this research.

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Transfection Statistics from EGFP-Fluorescence Data

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Transfection of genetic material to cells by means of natural or artificial viruses is at the focus of modern gene therapy. The appropriate dose of medication is of vital interest: in opposition to direct medication with a vast number of drug molecules, one here tries to influence the metabolism in a top-down approach by inserting small numbers of genes, from which expression is initiated.

Since small numbers are involved, fluctuations gain importance, influencing the efficiency of medication. Here we present theoretical analysis what kind of protein distributions are to be expected and how conclusions can be drawn on plasmid number distributions.

If protein degradation is much slower than mRNA kinetics and the expression factor is large, the protein distribution shows multiple peaks, reflecting integer gene copy numbers. Experiments suggest that copy numbers are often very small. Successful expression initiations can be regarded as independent events, hence giving rise to a Poisson distribution of expressing plasmids. This distribution can be determined and so can the overall gene expression factor.

Morphological Control in Organic Electronic Devices via Block Copolymer Self-Assembly

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Morphology plays a crucial role in organic semiconducting devices. In organic solar cells, for example, one pivotal parameter is the creation of a large donor-acceptor interface for efficient charge separation while providing charge transport pathways to the respective electrodes at the same time. Here, we present the

synthesis [1] and application of novel donor-acceptor diblock copolymers that allow tuning of nanostructures to fulfil the requirements mentioned above. The morphology of these molecules is investigated by transmission (TEM) and scanning electron microscopy (SEM). Depending on the block lengths, these block copolymers self-assemble into elongated electron-conducting worm-like or lamellar nanostructures embedded in a hole-conducting matrix. Single layer polymer solar cells with only the block copolymer as active material reveal a very strong morphological dependence on the device performance [2].

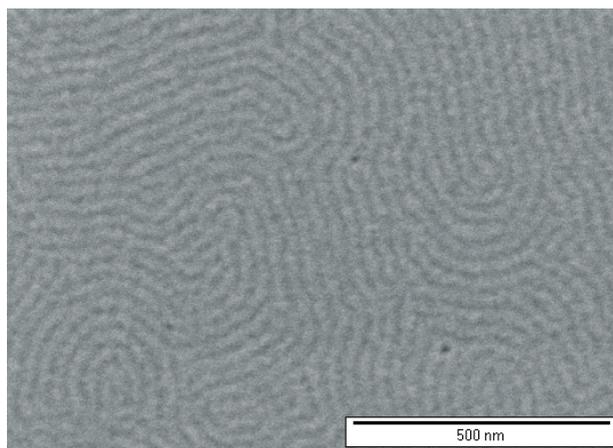


Fig. 1 Scanning electron micrograph of a thin film organic field effect transistor made of the novel electron-conducting block copolymer. 15 nm thin electron-conducting cylinders are observed as bright domains.

Furthermore, we will present a novel electron-conducting block copolymer that self-assembles into a perfect cylindrical morphology as evidenced by SEM and TEM. Organic field effect transistors with this block copolymer exhibit the same charge carrier mobility compared to a device made of the electron-conducting homopolymer. An explanation for

this exciting result is given by the morphological characterization of the OFET device. There, lying cylinders with a statistical orientation are observed which can serve as conduction pathways (Figure 1).

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Nanocrystal-Encoded Microbeads for Diagnostics of Autoimmune Diseases

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Early discovery of small amounts of autoantigens is essential for treatment of autoimmune diseases. A lot of effort is currently invested in development of fast and standardized detection schemes. In our work [1] we followed the fluorescence labelling approach, which offers advantages of rapidity, objectivity and potential for automation.

Polymer microbeads were optically encoded with fluorescent CdSe/ZnS nanocrystals and used for a systemic sclerosis flow cytometry immunoassay. The microbeads were prepared using electrostatic layer-by-layer deposition of polymers in combination with infiltration of nanocrystals. Surface of the beads was covered with fragments of human DNA topoisomerase I (topoI). Anti-Scl-70 (antitopoisomerase I) antibody, systemic sclerosis marker, bound to the surface of topoI covered the microbeads. A dye labelled secondary antibody to anti-Scl-70 was used as anti-Scl-70 reporter. Flow cytometry data plots allow to distinguish between the samples with and without the analyte. The achieved sensitivity was less than 1 pmol of autoantibodies. Results of confocal microscopy and time-resolved FRET measurements support the formation of topoisomerase I - antitopoisomerase I - anti-antitopoisomerase I immunocomplex. The presented immunoassay is the first application of nanocrystal encoded microbeads in proteomics.

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Nanopatterned Surfaces by Molecular Self Assembly - a STM Study

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In the last decades top down engineering via lithographic methods led to high performance functional devices. But it is predicted that the now used common methods will reach their limits at structures below 20 nm. Contrary self assembly is a powerful method to obtain even smaller defined functional structures and has found use in miscellaneous fields such as chemical sensors [1], biotechnology [2], or catalysis and electrochemistry [3]. Self assembled monolayers of metal organic compounds - especially imine bridged complexes - are very interesting because of their catalytical - and in the case of compounds bearing two or more metals - of their paramagnetic properties.

Here we describe the synthesis of a self organizing ligand systems based on 2,4-dihydroxybenzaldehyde [4] and 2,4-dihydroxyacetophenon as well as the corresponding mono- and binuclear complexes. Furthermore derivatives of gallic acid bearing long alkyl chains and UV sensitive units were prepared. The monolayers of the named compounds were investigated at the liquid/HOPG interface using scanning tunneling microscopy. The design of the ligand systems led to highly ordered lamellar 2D structures which are dependent on the alkyl chain length and on the metals contained. The assembly of monolayers is driven by attractive van der Waals interactions of the alkyl chains and occurring H-bonds between neighbouring molecules. Thereby the array of the pure ligands can easily be distinguished from the pattern of the corresponding metal complexes.

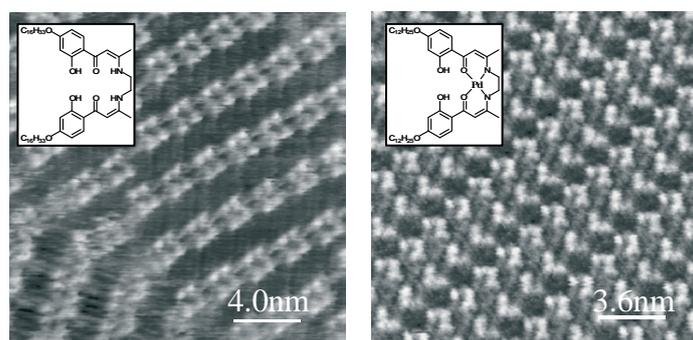


FIG. 1 STM image of an Extended Salen ligand and its corresponding Pd-complex

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Sequence-dependent Fluorescence of DNA-hosted Ag Nanoclusters

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We study how the fluorescence of DNA-bound silver nanoclusters depends on the DNA strands that host the Ag atoms. Comparison of the visible fluorescence produced by Ag bound to single-stranded DNA and to double-stranded DNA shows that Watson-Crick base pairing suppresses formation of optically active Ag-DNA species. For single-stranded DNA, the wavelengths of the primary fluorescence peaks are highly reproducible and sensitive to the DNA sequence. Immobilization by drying enables the imaging of single emitters, as evidenced by the observation of dipolar emission patterns and fluorescence intermittency. The sensitivity of the Ag-DNA fluorophores to the DNA sequence and the straightforward detection of single emitters hold promise for future applications in nanoscale DNA-based devices.

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Electret Additives for Isotactic Polypropylene

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In order to improve the charge storage properties of isotactic polypropylene (i-PP) several trisamides with different substituents and varying cores have been incorporated. When added in appropriate concentrations, these additives act as charge traps by the formation of isolated nano-aggregates and thus inhibit charge drift and neutralization [1]. This work focuses on the establishment of structure – property relations, as well as on the concentration dependence.

The additives have been incorporated into i-PP by extrusion, and 80 μm thick films were made via compression molding of injection molded specimens. The films were charged using a point-to-plate corona setup with a grid for limitation of the surface potential. The surface potential of the samples was measured with an electrostatic voltmeter. The measurements were performed directly after charging at room temperature to get the initial value for the surface potential and after annealing at 90 °C for cumulative periods of 30, 90, 180, 360, 540, and 1440 minutes resulting in a surface potential decay curve. The high annealing temperature is used to simulate long-term measurements at room temperature.

Exemplarily, the surface potential decay curves of compound 1a is shown in five different concentrations in comparison to a reference compound 2, that is not able to build intermolecular hydrogen bonds, and in comparison to neat i-PP (Fig. 1). The optimal concentration of 1a in i-PP is 0.013 wt%, whereby the charge retention is about 50 Volt higher than that of the unadditivated i-PP. The presence of compound 2 makes the surface potential drop down clearly.

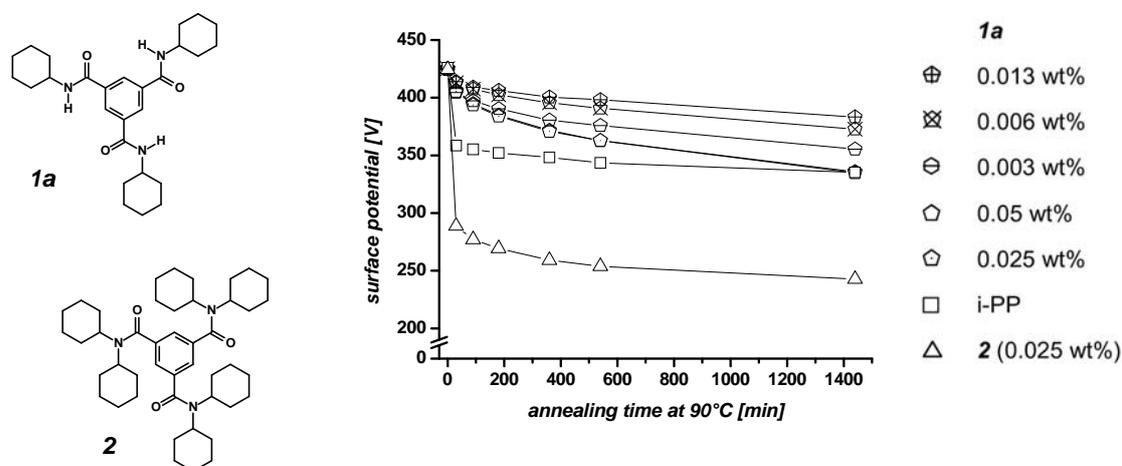


Fig. 1 Surface potential decay curves of benzene-1,3,5-tricarboxylic acid-(N-cyclohexyl)-trisamide (1a) in different concentrations, benzene-1,3,5-tricarboxylic acid-(N,N-dicyclohexyl)-trisamide (2), and neat i-PP.

This work was supported by the DFG (Projekt Schm 703/2-2)

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Fluorene Based Reactive Mesogens for Photopatterning

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Semiconducting organic materials have gained interest in the last years due to their applications in displays, electronics or sensors. Especially organic light-emitting diodes (OLEDs) are currently emerging as promising technology for flat panel displays and large area lighting [1,2]. In view of the mentioned applications patterning techniques for the production of pixelated devices are of particular interest. Conventional photolithography techniques optimized for inorganic materials are difficult to apply to organic materials. It was shown that classic photoresist techniques can only be transferred to structure organic compounds with major modifications, e.g. as lift-off techniques [3], which limits their applicability in OLED manufacturing.

Thus an attractive approach is the direct photochemical crosslinking of the semiconducting material itself, for example by the introduction of photocrosslinkable moieties [4,5]. By light induced crosslinking the material becomes insoluble and can simultaneously be patterned, comparable to a negative photoresist.

Here we report the synthesis, characterisation and photocrosslinking of fluorene based reactive mesogens. Polyfluorenes are well known emitter materials for organic light emitting devices due to their strong blue fluorescence, high quantum yields and easy handling.

Compounds bearing different contents of crosslinkable sites were synthesised and subsequently the photopolymerisation was examined. Under optimized conditions μm -sized patterns were obtained (see Fig.1).

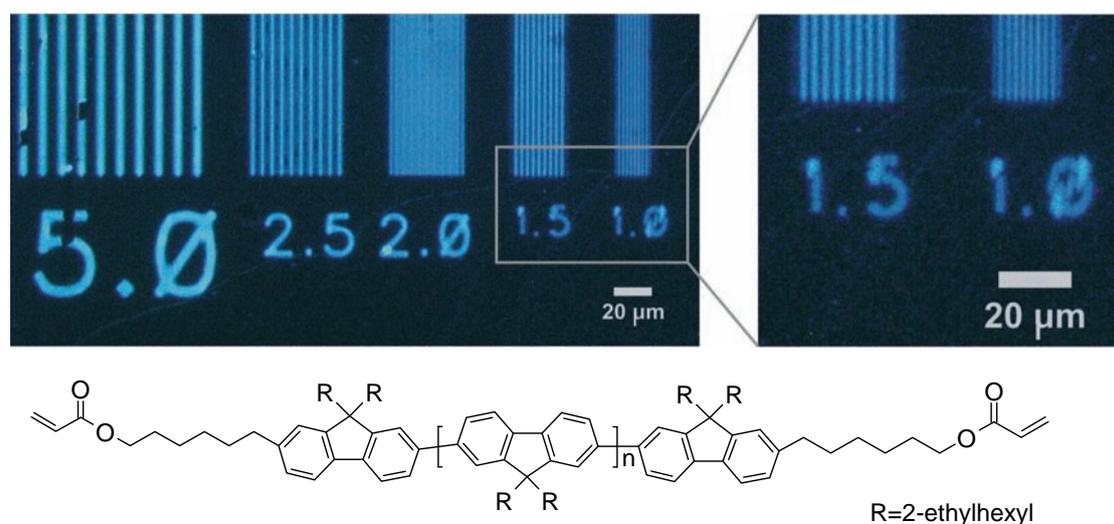


Fig. 1 Fluorescent microstructures (observed under a fluorescence microscope) of the depicted oligofluorene (1 wt% photoinitiator, irradiation with UV light, development in THF).

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The mechanism of Gene Regulation Revealed by SpFRET

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Gene expression is an essential process in life and starts with the transcription of DNA into messenger RNA. The first step in the initiation of DNA transcription is the binding of the TATA-Box Binding Protein (TBP) to the gene promoter site on the DNA. The TBP recruits other transcription factors necessary for transcription and is a primary target for gene regulation. The transcription can be either enhanced or inhibited by regulatory proteins, such as Negative Cofactor 2 (NC2). NC2 forms a ring complex with the DNA bound TBP and represses gene expression in vitro [1,2]. Recent results of biochemical experiments suggest that the TBP becomes mobile along the DNA upon the binding of NC2. Thus, inhibition by NC2 could be explained by the dynamic behavior of TBP rather than by steric hindrances as currently thought.

To test this hypothesis, we have performed fluorescence resonance energy transfer (FRET) experiments on single TBP-NC2 complexes bound to DNA using Total Internal Reflection Fluorescence Microscopy with dual color detection. TBP was labeled specifically with a FRET-donor molecule and a 70 bp dsDNA containing a TATA sequence in the middle of the strand was labeled with a FRET-acceptor. The DNA was immobilized on the surface of the sample chamber. Before addition of NC2, the TBP-DNA complexes displayed a steady FRET signal. Upon addition of NC2, fluctuations in the FRET efficiency of individual complexes were observed. The variations in FRET occur from changes in the conformation of the DNA as well as movement of the TBP-NC2 complex along the DNA and have been observed in experiments using several different promoters.

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Coagulation Kinetics of Spherical Polyelectrolyte Brushes in Presence of Multivalent Counterions

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In this work, we studied the coagulation kinetics of a monodisperse colloidal spherical polyelectrolyte brush (SPB) in the presence of multivalent Lanthanum(III) counter ions. The particles used here consist of a polystyrene core of 115 nm radius onto which linear poly(styrene sulfonate) (PSS) chains are chemically grafted via a photo-emulsion polymerisation. This procedure led to a dense brush of PSS chains with a contour length L_c of 48 nm. Determination of coagulation kinetics was performed by time-resolved simultaneous static and dynamic light scattering in the early stage of coagulation as described by Holthoff *et al.* [1]. Time-resolved simultaneous static and dynamic light scattering permitted the determination of the absolute coagulation rate from the change of light scattering intensity as well as from the increase of the hydrodynamic radius without knowledge of light scattering form factors of the aggregates. The fast coagulation rate constant of the SPB in Lanthanum(III)chloride solution was found to be $3.9 \pm 0.2 \cdot 10^{18} \text{ m}^3/\text{s}$. A stability curve was measured using different Lanthanum(III)chloride concentrations in the range of 0,2 mM to 0,15 M. We proposed a new theoretical model of the inter particle interactions of the SPB in presence of multivalent counter ions. This model describes the overall interaction pair potential assuming a strong attractive interaction upon direct contact of the particles and an electrostatic repulsive potential at larger distances. By comparing the theoretical model with the experimental data, we concluded a linear decrease of the surface potential of the SPB with increasing Lanthanum(III)chloride concentration.

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Imaging SNARE Mediated Membrane Fusion Events with Single Vesicle Resolution

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SNARE (soluble N-ethylmaleimide-sensitive-factor attachment protein receptor) proteins seem to mediate intracellular membrane fusion in all secretory pathways in eukaryotic cells [1]. According to the SNARE hypothesis [2], each type of transport vesicle has a distinct v-SNARE that pairs up with a unique cognate t-SNARE at the appropriate target membrane [3]. Here we present a fluorescence assay with single vesicle resolution for the quantitative study of docking, fusion and hemifusion events. Planar lipid bilayers containing binary t-SNAREs (syntaxin-1A associated with SNAP-25) were incubated with proteoliposomes reconstituted with v-SNAREs (VAMP-2) [4].

We found that variation of the receptor surface density from 0–5,000 t-SNAREs/ μm^2 yielded densities of adsorbed vesicles ranging from 0.1–0.4 v-SNARE vesicles/ μm^2 and that docking is extremely sensitive to surface impurities. Impurity effects result in massive docking and can overcome the receptor concentration dependency. Membrane fusion (see Fig. 1) took place at fusion rates of $2\text{--}4\cdot 10^{-4} \text{ s}^{-1} \mu\text{m}^{-2}$ on surfaces containing t-SNARE receptors in low concentrations (~ 60 t-SNAREs/ μm^2). High receptor concentrations ($\sim 4,000$ t-SNAREs/ μm^2) resulted in a complete suppression of fusion. The addition of $100 \mu\text{M}$ Ca^{2+} did not increase the fusion rate. We observed fusion $0.11\text{--}1.1$ s after docking of the vesicles depending on the vesicle and surface receptor concentration. Hemifusion is less frequent than complete fusion and occurred generally after 0.69 s. Vesicle fusion is immediately followed by outward radial diffusion of fluorescent lipids with a diffusion coefficient of $D = 0.3 \mu\text{m}^2 \text{ s}^{-1}$.

The method we developed should allow for further investigations of the interesting and unexpected findings in order to build a more comprehensive picture of SNARE mediated membrane fusion.

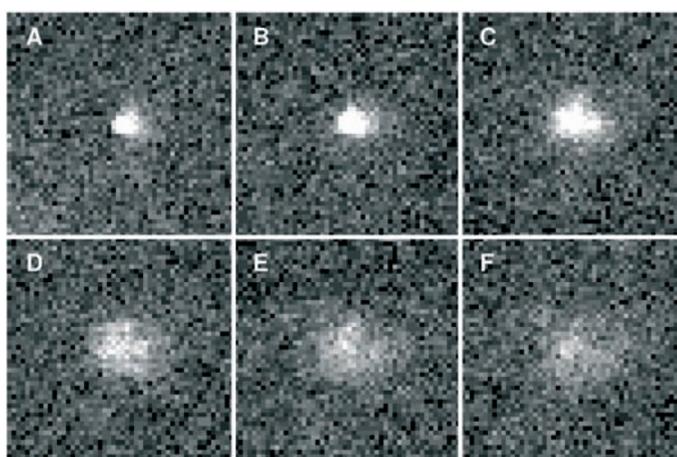


Fig. 1 Detection of fusion events by fluorescence microscopy. A) A vesicle docked onto the bilayer is seen as a sharp spot of fluorescence intensity. B) The onset of fusion is determined as an increase in the fluorescent area and the total fluorescence intensity. C,D) Fusion is accompanied by a radially extending cloud of fluorescence intensity. E,F) After several frames, the fluorescence intensity begins to disappear due to photobleaching of the diffusing fluorophores.

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Pulling on Siloxane Molecules

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Increasing the mechanical stability of artificial polymer materials is an important task in materials science and for this a profound knowledge of the critical mechano-elastic properties of its constituents is vital. Here, we use AFM based single-molecule force spectroscopy measurements to characterize the rupture of a single silicon-oxygen bond in the backbone of polydimethylsiloxane as well as the force-extension behavior of this polymer. PDMS is not only an elastomer used in a large variety of products, but also an important model system for highly flexible polymers. In our experiments we probe the entire relevant force range from low forces dominated by entropy up to the rupture of the covalent Si-O bonds in the polymer backbone at high forces. The resulting rupture-force histograms are investigated with microscopic models of bond rupture under load and compared to density functional theory calculations to characterize the free energy landscape of the Si-O bond in the polymer backbone.

With this information it is possible to investigate the forces holding single polymers in complex networks. These polymer films consist of a rigid catalytical interlinked host polymer and a guest polymer which is held in the host polymer only by entanglement interactions. Additional filler particles can be introduced into the sample. These interact with both the host- and the guest polymer to change the anchoring of the guest polymer in the network.

Hydrogen Production by the Photo-cleavage of Water using TiO₂ Nanotube Arrays

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Vertically oriented Titania Nanotubes grown by anodization constitute a material architecture that can be precisely tuned to maximize light absorption, charge separation and charge collection. Thus, electronic carriers can be generated in this architecture by incident light in sufficient numbers and then efficiently used to perform work, rather than simply recombining. Titania Nanotubes offer a technology that can be beneficially used to degrade organic pollutants, split water to generate hydrogen, generate electricity from anchored dye molecules and decontaminate water (as a germicide) under the action of sunlight and/or ultraviolet radiation. Dye sensitized titania nanotube photoelectrodes have been shown to convert sunlight into electricity with an efficiency of 7 %. Under band-gap illumination, 30 µm long titania nanotube array based photoanodes performed water-splitting at an efficiency of 16.25 %.

In the last year, we have had a number of significant breakthroughs relating to our work on titania nanotube arrays. The first relates to the length of the nanotube arrays that can be grown by anodization. By using polar organic electrolytes such as formamide and ethylene glycol in the anodizing electrolyte, the maximum length of the nanotube arrays has been increased from 7 µm to 700 µm. Also, Ti-Fe-O nanotube arrays were successfully fabricated from cosputtered films of titanium and iron. Films with an iron content of 6.2 % exhibited the best photo-electrochemical properties. The Ti-Fe oxide nanotube array films were utilized in visible spectrum water photolysis, demonstrating a photocurrent of 2 mA/cm² under AM 1.5 illumination with a sustained, time-energy normalized hydrogen evolution rate by water splitting of 7.1 mL/W•hr in a 1 M KOH solution.

From DNA-based Nanotubes to DNA-wrapped Carbon Nanotubes

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DNA is proving to be a suitable and useful building block at the nanoscale. Its primary advantage is the programmability of structure and size. We are using DNA to assist in templating the arrangement of nano-sized objects such as fluorescent molecules, gold nanoparticles, and quantum dots. Here we present information about the design and experimental analysis of two different structures which will help us do this: DNA-based nanotubes and DNA-wrapped carbon nanotubes. These may prove suitable for use in biosensors and fundamental studies of collective phenomena.

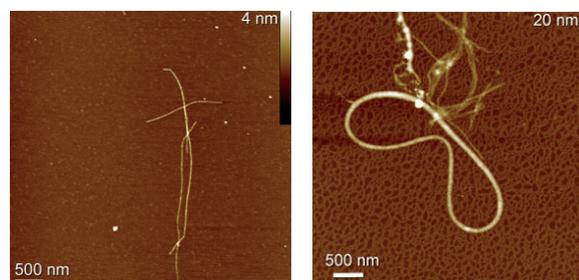


Fig. 1 Atomic Force Microscope topographic images of: a) DNA-wrapped carbon nanotubes. b) DNA nanotube.

Compartmentalized Cylindrical Nanoparticles – Preparation, Solution Properties and Side-Specific Decoration with Inorganic Nanoparticles

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Block copolymers offer a wide variety of well-defined microphase segregated structures in the bulk phase. This contribution reports on the preparation of cylindrical nanostructures with internal compartmentalization. The particles are prepared via crosslinking the polybutadiene segments of different morphologies of linear triblock copolymers and a miktoarm star polymer. Due to the unique morphologies found in the bulk state, different kinds of cylinders can be obtained. For instance, crosslinking a polystyrene-*block*-polybutadiene-*block*-poly(methyl methacrylate) triblock terpolymer, it is possible to obtain Janus cylinders. These cylinders possess a crosslinked core of polybutadiene and two hemicylinders as corona (PS, PMMA).

An even further subdivision of the corona of the cylinders can be achieved by changing the polymer architecture from linear to a heteroarm star polymer, bearing arms of polybutadiene, poly(2-vinylpyridine) and polystyrene. This leads to the formation of new morphologies, which cannot be obtained for linear terpolymers. The cylinders obtained on basis of the heteroarm star polymer template exhibit a corona with four segments, two PS and two P2VP. The segments oppose each other and the complete structure is centrosymmetric. Figure 1 shows the schematic synthesis and the side-selective decoration of the P2VP segments with gold nanoparticles.

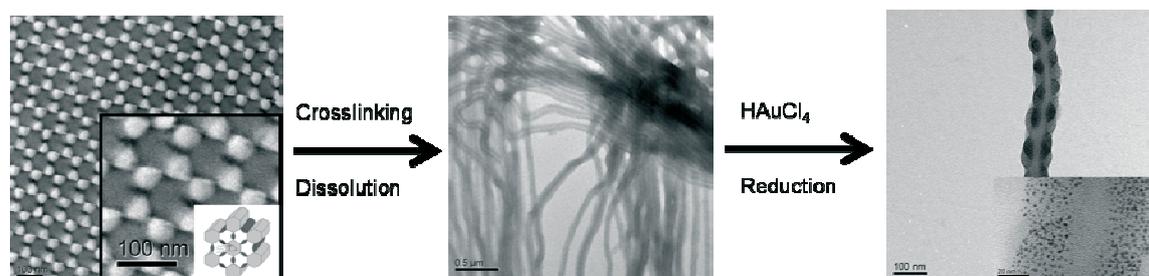


Fig. 1. Schematic Synthesis of multicompartment cylinders on the basis of a heteroarm star polymer and its side-specific decoration with gold nanoparticles.

Results concerning size evolution, solution properties and aggregation tendency are presented. The results were obtained using complementary imaging and scattering techniques. In addition, side-specific decoration of the different compartments with inorganic nanoparticles will be presented, rendering these materials interesting for potential applications as novel nanodevices.

Frequency Modulation Torsional Resonance Mode AFM (FM-TR-AFM) for the In-plane Surface Properties Measurement

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Torsional vibration of atomic force microscope (AFM) cantilevers can be used to measure elastic and friction properties on a nanoscale. The vibrations of the cantilever in the torsional mode are excited by two piezoelectric elements attached beneath the holder of the chip, in which two piezo actuators vibrate out-of-phase with respect to each other [1]. The torsion about the long axis of the cantilever promotes a small pendulum-like oscillation of the tip apex parallel to the sample surface with typical amplitudes of 0.2nm to 2.0nm. This pendulum-like oscillation parallel to the surface avoids the strong nonlinearity due to the vibro-impact dynamics of conventional methods such as tapping or non-contact mode.

In this work, we demonstrate frequency modulation (FM) for torsional resonance mode AFM. Both, frequency shift and energy dissipation are measured under ambient conditions. The frequency shift provides information on the conservative interaction, while the dissipation signal contains information on additional energy losses. To regulate the FM detection scheme, we used a Phase-Loop-Look controller/detector (PLL) from Nanosurf (Liestal, Switzerland). By monitoring the changes in the resonance frequency and the excitation amplitude required to keep the oscillation amplitude at a constant value, we were able to measure the frequency shift and dissipation caused by the tip-sample interaction. Frequency shift tip-spacing curve taken on homopolymers polystyrene (PS) reveals that the different chain length of the polymer surface shows different response to the conservative and the dissipative tip-sample interaction. The distance between the first contact between tip and surface and the transition from the interfacial layer to the bulky polymer corresponds very well to the radius of gyration of the PS homopolymers investigated here [2]. The frequency shift versus displacement curves can be used to differentiate the different polymer surface, from their molecular weight.

The demonstration the energy dissipation mapping by frequency modulated torsional resonance AFM under ambient conditions opens a wide field of new applications in tribology. Frictional properties can be addressed by dynamic AFM which have not been accessible by the NC-AFM in vertical direction to the sample surface so far.

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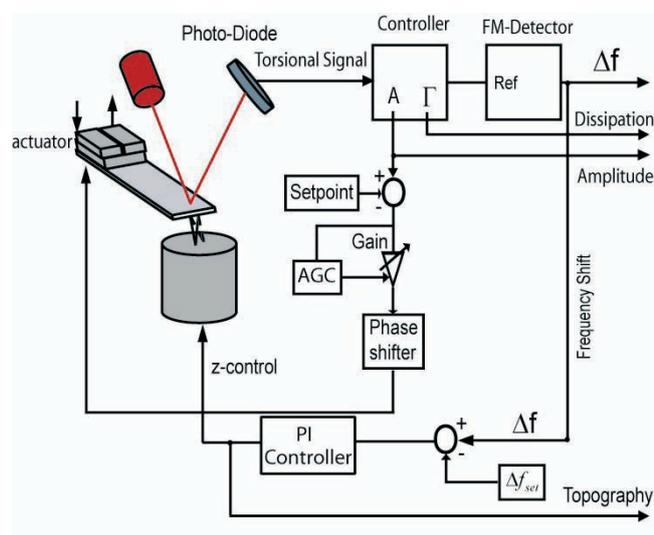


Fig. 1 Schema of the feedback circuitry used for frequency modulation of torsional resonance imaging. The torsion signal is measured by the quadrant photodiode. The signal was pre-amplified and then fed into the FM-Controller. The frequency shift signal was used for feedback and the damping was detected as amplitude variation by the automatic gain control (AGC).

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	Thursday, 06.09.07	Friday, 07.09.07	Saturday, 08.09.07	Sunday, 09.09.07
07:30		Breakfast	Breakfast	Breakfast
09:00	Departure from Munich at 10:30 Arrival in Irsee at ~12:00	Design of polymer-coated nanoparticles for positioning in block copolymers <i>Ed Kramer</i>	Nondisruptive block copolymer-based nanolithographic processes <i>Thomas Russel</i>	Complementary coordination of porphyrin as scaffold for biomimetic and optics materials <i>Yoshiaki Kobuke</i>
09:45		Surface-Confined Self-Assembly <i>Mario Ruben</i>	Can an optical sensor for TNT related explosives be made? <i>Jan O. Jeppesen</i>	Multifunctional donor-acceptor block copolymers for solar cell applications <i>Mukundan Thelakkat</i>
10:30		Break	Break	Break
11:00		Correlation between chain morphology and electronic structure in conjugated polymers <i>Enrico Da Como</i>	Controlling complex fluids in microsystems <i>Stephan Herminghaus</i>	DNA-switchable supramolecular structures <i>Fritz Simmel</i>
11:45		Micromechanics of ultrathin polymeric membranes - from capsules to wrinkles <i>Andreas Fery</i>	Slow transport in complex environments <i>Thomas Franosch</i>	CLOSING
12:30		Lunch (Buffet)	Lunch (Buffet)	Lunch (Buffet)
14:15	OPENING			
14:30	DNA as a tool for Nanoscience <i>Deborah Fygenson</i>	Synthesis and application of vertically oriented, highly ordered arrays of TiO ₂ nanotubes <i>Craig Grimes</i>	Cell-free gene expression on a chip <i>Roy Bar-Ziv</i>	
15:15	Wormlike and glassy wormlike chains <i>Klaus Kroy</i>	Short presentations of posters I	Revealing the interplay of forces: dewetting fluids, adsorbing proteins and sticking geckos <i>Karin Jacobs</i>	
16:00	Break	Break	Break	
16:30	Tailor-made surfaces for microsystems and nanotechnology <i>Jürgen Rühle</i>	Short presentations of posters II	Nanostructures for Photovoltaics <i>Lukas Schmidt-Mende</i>	
17:15	Tuning block copolymer morphologies with electric fields – from chain stretching to order-order-transitions <i>Alexander Böker</i>	Poster Session	Block Copolymer Blends for Holographic Data Storage <i>Hans-Werner Schmidt</i>	
18:00	Dinner (Buffet)			
19:30		Dinner (Buffet)	Conference Dinner (Menue)	