

CPP 15: Single Molecules

Time: Tuesday 17:00–19:00

Location: C 230

CPP 15.1 Tue 17:00 C 230

Single molecule probes in soft materials — ●JÖRG SCHUSTER and CHRISTIAN VON BORCZYKOWSKI — TU Chemnitz, Institut für Physik, 09107 Chemnitz

Within the past years, single molecule spectroscopy has become a routine tool which is extensively used to study soft, mostly biological, materials. The success of single molecule spectroscopy is due to the fact, that single molecule probes are sensitive to the structure and dynamics of their local nanoscale environment. Fluctuations of single molecule observables such as dipole orientation, spectral positions, excited state lifetime which would be averaged out in ensemble experiments are exploited in order to detect static and dynamic heterogeneities in combination with a nano scale resolution.

The present talk will be focussed on the application of single molecule probes in the field of nonbiological, soft materials, including simple and complex liquids as well as polymers. A number of very different single molecule based techniques is used, including single molecule tracking in a wide field microscope as well as spectroscopy in a confocal instrument. The application of special dye probes will be demonstrated by some recent experiments from our group. In addition, we report on a very recent finding, namely to use the so called power law intermittency (blinking), as a probe of local dielectric relaxations and charge trapping in nonconducting polymer materials.

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Molecular Wires in Single Molecule Junctions: Charge Transport and Vibrational Spectra — ●STEFAN BALLMANN¹, DANIEL SECKER¹, HEIKO B. WEBER¹, WOLFGANG HIERINGER², and ANDREAS GÖRLING² — ¹Lehrstuhl für Angewandte Physik, Universität Erlangen-Nürnberg, Staudtstrasse 7, 91058 Erlangen — ²Lehrstuhl für Theoretische Chemie, Universität Erlangen-Nürnberg, Egerlandstrasse 3, 91058 Erlangen

Quantum transport through single molecules is accompanied by internal molecular vibrations, detectable in principle as vibronic side-peaks in the conductance spectra. We use the mechanically controllable break-junction (MCBJ) technique for the investigation of single-molecule contacts. The molecules under investigation are oligoynes, with a string-like atomic chain unit consisting of eight carbon atoms with alternating single and triple bonding, spanned between two platinum atoms and protected by bulky ligands. In our experiments, suppressed conductance at low bias, characteristic step-like features at higher voltages and strong sample-to-sample fluctuations are observed in the I - V -characteristics. In order to obtain a better understanding of the experimental results, density functional theory (DFT) calculations were performed. The experimental data show strong indications for the observation of vibrational features as peaks in dI/dV at a temperature of 75 Kelvin. In comparison to the theoretically observed values for the vibrational frequencies of the carbon chain, the peaks are in excellent agreement with the four energetically lowest-lying longitudinal modes.

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Numeric force ramp spectroscopy for polymer stretching: the role of thermal fluctuations — ●FELIX HANKE^{1,2}, DOUGLAS B STAPLE², and HANS JÜRGEN KREUZER² — ¹Fritz-Haber Institut der MPG, Faradayweg 4-6, 14195 Berlin — ²Dept of Physics, Dalhousie University, Halifax, NS, B3H 3J5, Canada

Force ramp spectroscopy is aimed at stretching single polymer molecules in an Atomic Force Microscope (AFM) such that the external force applied to the molecule increases linearly. This approach differs from the more commonly used constant velocity experiments in the observed thermal noise of the molecule-cantilever system. Recent experiments on Dextran [1], doubted the applicability of the often-used Gibbs (or fixed force) ensemble based on a thermal fluctuations argument. Here we use a Transfer Matrix formalism to describe the Dextran-cantilever system [2], which is coupled to a master equation description of the pulling process [3]. This theory enables a numerical imitation of the force ramp experiment from which the molecular force response and thermal noise of the force ramp setup are calculated. We find that the thermodynamics governing the force ramp experiment always depends significantly on the AFM cantilever. Specifically, the thermal noise of the system always remains restricted by the cantilever.

This leads us to suggest that the interpretation of every single-molecule AFM experiment must account explicitly for the effect both, molecule and cantilever. [1] Walther et al, Biophys J 90 3806 (2006); [2] Hanke, Kreuzer, Eur Phys J B, 22 163 (2007); [3] Hanke, Kreuzer, Phys Rev E, 72 031805 (2005)

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Time-resolved photoluminescence studies on individual single carbon nanotubes — ●TOBIAS GOKUS¹, HAYK HARUTYUNYAN¹, FRANK HENNRICH², MANFRED KAPPES², and ACHIM HARTSCHUH¹ — ¹Department Chemie und Biochemie, Ludwig-Maximilians-Universität München and CeNS, D-81377 München — ²Forschungszentrum Karlsruhe, Institut für Nanotechnologie, D-76021 Karlsruhe

The photoluminescence lifetime of individual semiconducting single-walled carbon nanotubes (SWCNTs) dispersed on glass substrates has been measured using time correlated single photon counting (TCSPC) at room temperature. We observe mono-exponential decay dynamics of the exciton recombination over more than four orders of magnitude in agreement with previous measurements at low temperature [1]. All photoluminescence transients show fast decay times in the range of few picoseconds (~ 10 ps) with large variations from nanotube to nanotube of the same chirality (n,m). To clarify the origin of these lifetime variations we studied the effects of finite tube-length, excitation energy and nanotube environment for different nanotube chiralities.

[1] A. Hagen et al., Phys. Rev. Lett. 95, 197401 (2005).

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Unravelling single polymers on surfaces by scanning force microscopy manipulation — ●WEI ZHUANG¹, EDIS KASEMI², FIKRI E. ALEMDAROGU³, A. DIETER SCHLÜTER², ANDREAS HERRMANN³, and JÜRGEN P. RABE¹ — ¹Department of Physics, Humboldt University Berlin, Newtonstr. 15, 12489 Berlin, Germany — ²Institute of Polymers, Swiss Federal Institute of Technology, ETH-Hönggerberg, HCI J 541, 8093 Zürich, Switzerland — ³Max Planck Institute for Polymer Research, Ackermannweg 10, 55128 Mainz, Germany

Scanning Force Microscopy (SFM) has become a powerful tool to reveal the superstructure of single polymer molecules as well as to manipulate individualized polymers on solid substrates into supermolecular functional entities, which would not form spontaneously. The manipulation can be brought about by either exerting a heterogeneous point force with an SFM tip in contact to a single polymer, or by a so called blowing manipulation with a tapping tip, which can exert a homogeneous force to the single polymer with circular topology. Here, we report an SFM tip manipulation of single dendronized polymers (denpols), which allows to determine the self-organized superstructure of denpols and can finally unravel the duplex superstructure of single charged denpols absorbed on solid substrate upon vacuum drying. In addition, we report an SFM blowing manipulation of single DNA-PEG-DNA triblock copolymer, so that the ds-DNA and the organic polymer chain have been unravelled against random coil, which afforded for the first time to visualize all three blocks of a single linear triblock copolymer chain with recognizable contours by SFM.

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Probing polymer dynamics by interfacial heat transfer at single gold nanoparticles — ●ROMY RADÜNZ and FRANK CICHOS — Molecular Nanophotonics Group, University of Leipzig, Linnéstr. 5, 04103 Leipzig

A new time-resolved photothermal microscopy technique has been developed, which allows to follow the release of heat from a single nanoparticle to the local surrounding. Photothermal microscopy is based on the conversion of light absorbed by a single nanoparticle into heat, which is released into the polymer matrix and induces a local refractive index change on a length scale on the order of 100 nm. Finally, this refractive index change can be detected optically with a confocal microscope by heterodyne optical techniques. Within this all-optical setup gold nanoparticles with a diameter down to 10 nm can be detected. By following the temporal evolution of the photothermal signal we are able to probe heat transfer across nanoscale interfaces between single metal nanoparticle and polymer matrix, that depends on the interfacial heat transfer resistance and the heat conductivity of the polymer. Furthermore, it is possible to probe thermal properties of the

polymer at various states of matter of the polymer. Temperatures even above melting temperatures of polymers are achievable due to the high absorption cross section and heat conversion efficiency of gold nanoparticles. The lengthscale on which these properties are probed can be controlled by applying an intensity modulated laser beam, since the heat diffusion length decreases with increasing modulation frequency.

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Mesopore functionalization as highly specific tool for the control of single molecule dynamics in silica materials —

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Nanoporous channel systems form a class of very promising host systems in many fields of modern science. Various guests can be embedded into their nanometer-sized pores. Yet, directly influencing the diffusion dynamics of the incorporated guest molecules is not an easy task, even though a broad range of applications (e.g. drug-delivery, catalysis) could profit from this ability. Covalently attached organic functionalizations within the porous network of the silica material could be an efficient tool for realizing this aim. Hence, this study examines the influence of different functionalizations onto the diffusion dynamics of single TDI dye molecules as guests within the mesoporous network of a hexagonal thin silica film. The films were synthesized with the non-ionic Block-copolymer Brij 56 as template. Within the pores the template micelles coexist with an organic carpet of functional groups. The specific choice of this functional groups then allows for a precise

tuning of the host-guest interactions and thus the guest dynamics. Furthermore this work will offer detailed mechanistic insights into the diffusion processes within a mesopore.

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Visualizing single molecule diffusion in nanoporous materials — •CHRISTOPHE JUNG, JOHANNA KIRSTEIN, ANDREAS ZÜRNER, BABARA PLATSCHEK, THOMAS BEIN, and CHRISTOPH BRÄUCHLE — Department of Chemistry and Center for Nanoscience (CeNS), Ludwig-Maximilians-Universität, Butenandtstrasse 11, 81377 München

Nanostructured host-guest materials are important for various applications (e.g. catalysis, molecular sieving, drug-delivery systems). Therefore a thorough understanding of the dynamics and the interactions of the guest molecules inside the host matrix is needed. Whereas measurements of ensemble diffusion provide information about the overall behaviour of the guests in a porous host, tracking individual molecules provides insights into heterogeneities, mechanistic details of molecular diffusion and the host structure. Thin mesoporous silica films containing a hexagonal and lamellar phase mixture were investigated. The dye molecules act as beacons while they diffuse through the different structural phases of the host: the structure of the trajectories, the diffusivities and the orientation of single molecules are distinctive characteristics, for molecules travelling in each mesophase or on the film surface. Moreover, we combine single molecule microscopy techniques with transmission electron microscopy in order to directly correlate individual molecular trajectories with the underlying structure of the porous host. It can be investigated in great detail how the luminescent molecule diffuses within the mesoporous channel system, how it changes speed, direction or traffic lanes in the porous network.