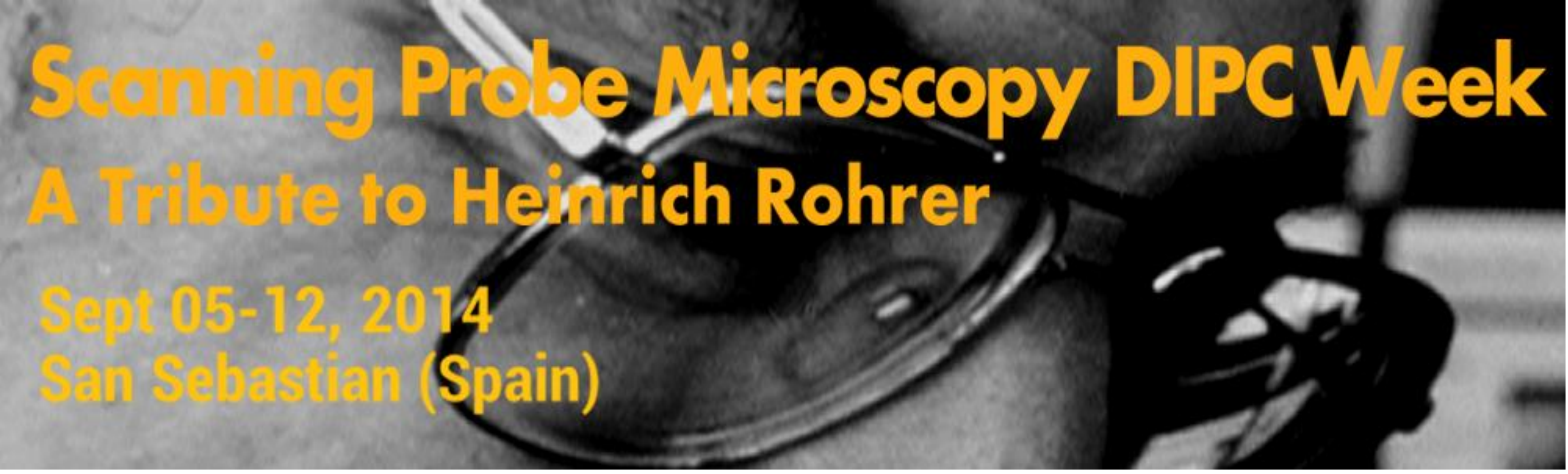

Scanning Probe Microscopy DIPC Week

A Tribute to Heinrich Rohrer

Sept 05-12, 2014
San Sebastian (Spain)

ABSTRACTS BOOK

Speakers and posters list - alphabetical order

Organisers

DIPC
Donostia International Physics Center

PHANTOMS foundation

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In collaboration with

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XXXIII Cursos de Verano
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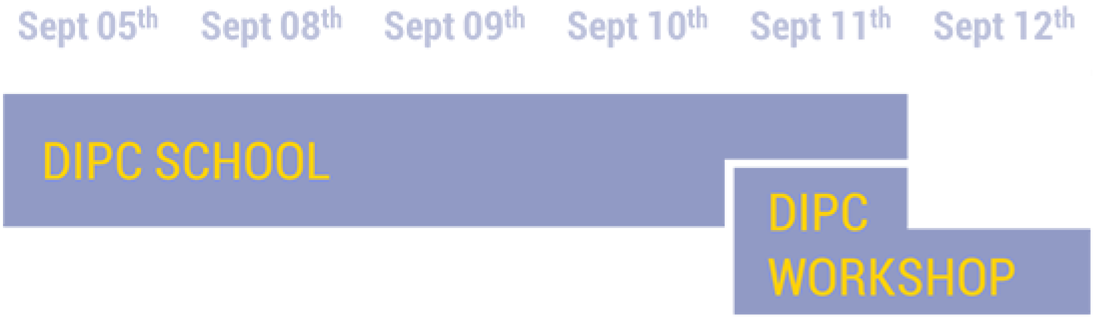
Europar Ikastaroak

The event, a Donostia International Physics Center (DIPC) School, is a tribute to Heinrich Rohrer (1933-2013) who shared the 1986 Nobel Prize in Physics with Gerd Binnig for the design of the scanning tunneling microscope (STM). This invention gave scientists the ability to image, measure and manipulate atoms for the first time.

The school will cover basic concepts and fundamentals of different SPM techniques including STM, Near Field Optical Microscopy (SNOM) and Atomic (AFM), Electrostatic (EFM) or Magnetic Force microscopes (MFM). It will also cover advanced topics such as time resolved SPM, density functional theoretical approaches, single molecule SPM or biology and nano-medicine applications.

These main topics will be complemented with a workshop with seminars on state-of-the-art SPM research, with participation of some of the most relevant scientist, who will give a first-person view of last discoveries on this field.



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ABSTRACTS

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Unusual Surface Faceting Induce by Metal Organic Complexes

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The actual demand of increasingly smaller devices drives the endeavors to explore new methods to miniaturize the designs. Thus, the development of methods capable of producing ordered nanostructured surfaces is a stimulating field.In this context, a novedous molecular/substrate interaction mechanism that derives in a unique adsorbate induce surface reconstruction is presented. In particular we show how metalated phthalocyanines can promote the formation of regular arrays of Cu nanoribbons on its (110) surface. At variance with the conventional changes of metal reconstructions upon molecular adsorption observed so far, the presented faceting is found to involve a massive reorganization of Cu adatoms. Thus, the energy gain of the final system comes not only from the preferential adsorption position of phthalocyanines on the copper surface, but also from their interaction with the sourrounded adatoms.
By combining experimental (Scanning Tunneling Microcopy) and theoretical surface science techniques we demostrate that indeed the mechanism behind the massive surface reshaping involves a molecular mediated uni-directional blocking of diffusing surface adatoms followed by their capture and accumulation.

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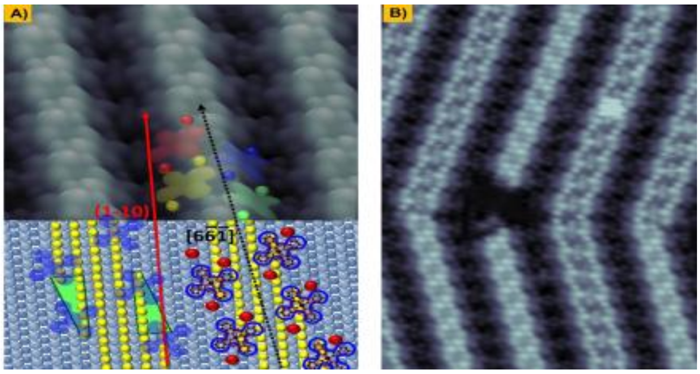

Controlling graphene plasmons with resonant metal antennas and spatial conductivity patterns

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Forthcoming information and communication technologies demand the manipulation of not only electrons but also optical fields at the nanoscale. A promising solution for an active control of light in such small regions is the excitation and manipulation of graphene plasmons, which offer ultra-short wavelengths, long lifetimes, strong field confinement, and tuning possibilities by electrical gating. The huge momentum mismatch between graphene plasmons and photons, however, presents a major technological challenge. Here, I will present a versatile platform technology that, based on resonant optical antenna structures (Fig. 1), allows for an efficient coupling of incoming light into propagating graphene plasmons. More importantly, I will show that these antennas and the use of spatial conductivity patterns (e.g. double layer graphene patches) also allow for controlling the graphene plasmons wavefronts, constituting an essential step for the development of graphene plasmonic circuits.

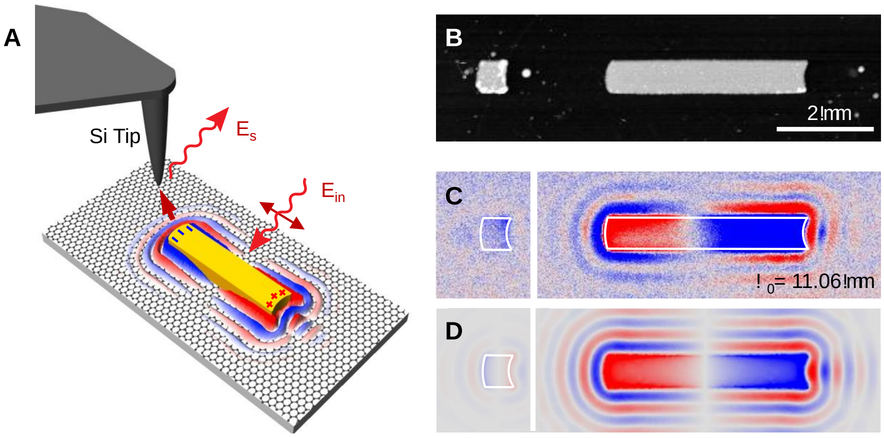

Figure1: Experimental verification of an antenna-based graphene plasmon device. (A) Illustration of the near-field imaging method. An illuminating plane wave with electric field Ein drives an antenna resonance in a metal nanorod. The subsequently generated near fields at the rod extremities launch plasmons in the graphene sheet, which covers the substrate. A dielectric Si tip scans the sample surface and scatters the local near fields at the sample surface. (B) Topography image of an off- and on-resonance dipole antenna (left and right, respectively). (C,D) Experimental near-field images, showing the real part of the vertical near-field component of the antenna shown in B.

Magnetic Force Microscopy

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Magnetism is one of the most important physical phenomena utilized by humans. The phenomenon of magnetic attraction has been know since ancient times because of the behavior of a variety of magnetite known as lodestone. Sometimes, magnetism has had a direct impact on human history, it was used for naval navigation already about 1000 years ago. Today, most of the total amount of recorded data is stored magnetically. Magnetic force microscopy (MFM) has been used extensively to characterize magnetic materials at nanoscale, i.e. recording media.

Magnetic Force Microscopy was developed in 1987 [1,2], only one year after the invention of atomic force microscopy, and since then applied to study domain structures in ferromagnetic thin films, small magnetic particles and artificially patterned structures. However, as MFM probes the stray field above the surface, its spatial resolution is limited to about 10 nm.

In this tutorial, we explore the fundamentals of this technique [3,4]. The particular material composition and shapes of the probes and the advantages and disadvantages of the different operation modes are discussed. The interpretation of images acquired by magnetic force microscopy requires some basic knowledge about the specific near-field magnetostatic interaction between probe and sample. Applications of magnetic force microscopy in the magnetic recording industry and in the fundamental research on magnetic materials are analyzed in terms of representative examples. An important aspect for any kind of microscopy is the ultimately achievable spatial resolution and the misinterpretation of the data in some particular cases.

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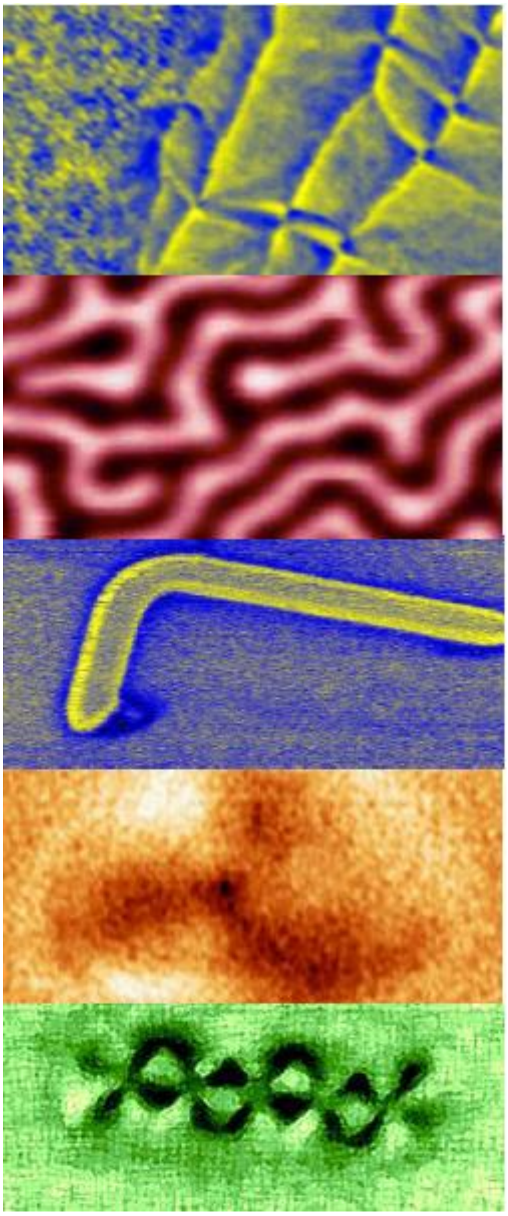
Figure



Dissipation in Magnetic Force Microscopy: artifacts or information?

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Measuring dissipation in scanning force microscopy relies on extracting physical information from either tip or sample by recording variations in the cantilever oscillation [1]. Non-conservative interactions between the tip and the sample give rise to a decrease in the oscillation amplitude of the lever, as some energy is transferred in every oscillation cycle into dissipative processes [2].

In this work, we analyze the useful information extracted from the dissipation in Magnetic Force Microscopy [3]. In some cases, the dissipation process is attributed to the rotations of the magnetization at the tip apex and that might be misinterpreted as part of the domain structure of the sample. We present a new way of mapping magnetic fields in three dimensions at the nanoscale, with a very high spatial resolution [4]. Figure 1 shows the Frequency shift (left column) and dissipation data (central column) compared to the simulations (right column), at increasing tip-sample distance. The profile along the black dashed line in the dissipation image at 100 nm, yields a FWHM of 7.6 nm. These experiments have been done in Py dots.

Furthermore, dissipation gradients in magnetic force microscopy have been found in materials with perpendicular anisotropy (i.e. CoNi multilayer films). The behavior of the dissipation versus the tip sample distance is analyzed [5]

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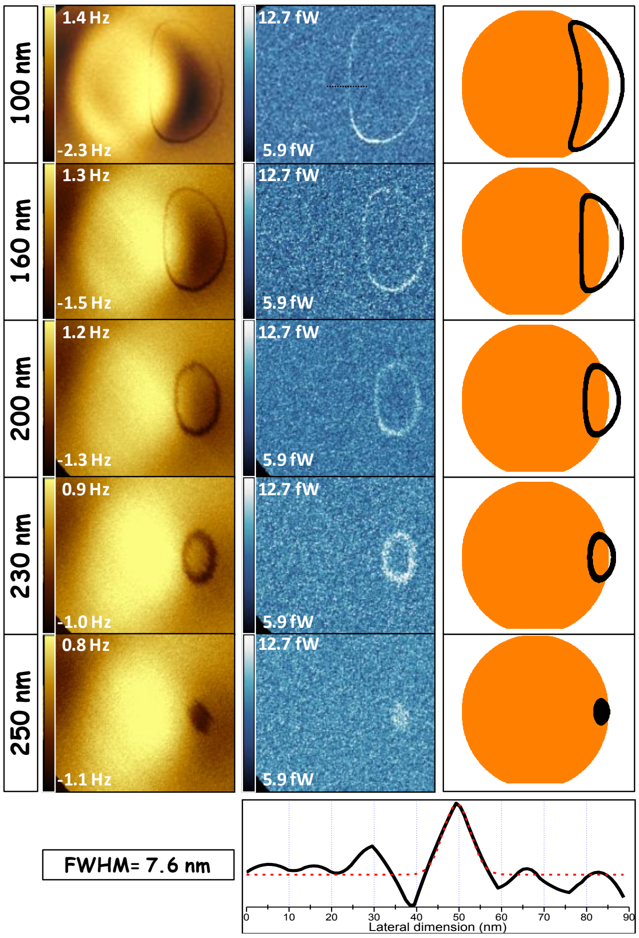
Figure


100 nm
160 nm
200 nm
230 nm
250 nm
1.4 Hz
-2.3 Hz
1.3 Hz
-1.5 Hz
1.2 Hz
-1.3 Hz
0.9 Hz
-1.0 Hz
0.8 Hz
-1.1 Hz
12.7 fW
5.9 fW
FWHM= 7.6 nm
0
10
20
30
40
50
60
70
80
90
Lateral dimension (nm)

Scanning Tunneling Microscopy at lowest temperatures: How to access the smallest energy scales

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As the energy resolution in a scanning tunneling microscope (STM) is principally connected to the temperature of the system, the smallest energy scales in solid state physics, such as the Zeeman splitting, the hyperfine splitting or elemental superconductivity, are only observable at lowest temperatures. With our new STM operating at 15mK, we have access to these small energy scales with an energy resolution of 11.4 ±0.3μeV. In this tutorial, I will address the challenges of reaching such low temperatures and what the limitations are. Using the Josephson effect in an STM tunnel junction, I will discuss in what way and to what extent the experimental setup becomes part of the measurement itself.

The Zeeman Effect in Confined Superconductors: A new probe of absolute spin polarization on the atomic scale

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When a dimensionally confined superconductor is subjected to a magnetic field, the compensating currents are suppressed such that superconductivity persists to much higher magnetic fields than in the bulk. This effect has been observed first in planar tunnel junctions by Meservey, Tedrow and Fulde [1]. The resulting Zeeman splitting of the quasiparticle density of states can be exploited as a probe for the absolute spin polarization of a spin polarized tunnelling current [2]. We have transferred this concept to the scanning tunnelling microscope (STM) to probe the spin polarization of the tunnelling current on the atomic scale by using a superconducting tip. As the superconducting tip carries no magnetic moment itself, the information in the spin polarized tunnelling current can be attributed to the magnetic properties of the sample. This is a decisive advantage over other spin-polarized STM techniques. First measurements on magnetic cobalt islands on a Cu(111) substrate are in excellent agreement with literature. Measuring the spin polarization of the tunnelling current as a function of tip-island distance, we find that the spin polarization increases by 65% when the distance is increased by only 2.3Å. This can be attributed to the different exponential decay of majority and minority states into the vacuum. This means that we can exploit the vacuum barrier as a tunable spin filter.

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Spin-polarization in real and reciprocal space: Analyzing the spin structure of electronic states by spin-polarized STM and quasi-particle interference

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The detection of spin-dependent surface properties is an important prerequisite for the understanding of various electron-correlation effects. Among the numerous techniques available, spin-polarized scanning tunneling microscopy (SP-STM) certainly has the highest resolution, as it is capable of imaging the magnetic spin structure of surfaces at ultrahigh resolution down to the atomic scale.

After revisiting the main features and achievements of SP-STM I will present recent results that demonstrate how the observation of interference patterns created by coherent electronic states, so-called quasi-particle interference (QPI), can be used to unravel the spin-polarization of electronic bands in k-space. In particular, lowtemperature scanning tunneling spectroscopy (STS) investigations of (√3×√3)- reconstructed surface alloys of some heavy post transition metals Ag(111), which provide giant Rashba-type spin-splittings, display some very complex QPI patterns. The analysis of the energy dispersion of the observed features and the comparison with density functional theory calculations shows how the conservation of spin angular and orbital momentum influences the interference pattern.

Light localization in near-field optics: theoretical foundations of scattering and propagation in the near-field

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This tutorial will develop fundamental concepts in scattering theory and their implications for near-field optics. We will cover the Green functions, evanescent fields, linear and nonlinear constitutive relations, the emergence of nonlocality from the microscopic theory, perturbative methods and applications in inverse scattering.

Nanoholography

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Scattering scanning near-field optical microscopy (s-SNOM) provides resolution of 5-10 nm and is inherently phase-sensitive. It is also slow, the bottleneck being the determination of phase and amplitude at each pixel. Holography is an efficient, multiplex method of acquiring phase across a whole image. I will report on the marriage of holography and s-SNOM, a new development that improves the speed of nanooptical imaging by orders of magnitude while simplifying the experiment. Examples will be given of large-scale (megapixel) imaging of manufactured nanostructures, CVD-grown materials, and the capture of dynamics not before possible.

Slow charge relaxation in (semi)-conducting polymers studied by Electrostatic Scanning Force Microscopy: Indication of Coulomb-Glass behaviour?

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The use of organic materials in optoelectronic devices promises to be a versatile alternative to traditional semiconductor technology. Correspondingly, organic electronics is a fast expanding field. The nanoscale morphology and opto-electronic properties of the basic building blocks of organic semiconductors -polymeric chains or complex molecules- is fundamental for their function [1]. In many cases, also the fundamental physical processes occur on a nanoscale. Scanning Probe Microscopy with its high resolution is thus an ideal tool for the study of this kind of materials.

In the present work Dynamic Scanning Force Microscopy is used to characterize the electronic properties of MEH-PPV and Polyalkythiophene thin films at high resolution. We apply Electrostatic and Kelvin Force Microscopy to these films and find surprising electrostatic properties of the films studied. First, the polymer morphology is not correlated with the electronic surface potential, and varies on a few minutes time scale (see Figure 1). Secondly, small surface potential domains (20-50nm), which are observed in the electrostatic signal have an interesting time evolution, which is again uncorrelated to the morphology, since the morphology is essentially static. The time evolution of the measured surface potential is studied using "movies" -that is the successive acquisition of images- of the sample were topography and surface potential are acquired simultaneously. We find on the one hand strong fluctuations of the surface potential and on the other a static mean corrugation of this potential. In addition to standard imaging experiments, to increase temporal resolution also "one-point" experiments were performed by fixing the lateral position (no scanning) and acquiring data at high speed at a specific location. These nanoscale local relaxation experiments show a slow relaxation with logarithmic time dependence over several decades [3]. Finally, not only electrostatic experiments were performed on these samples, also the time evolution of the surface photovoltage was studied using the recently developed "two pass" method (see [4] for further details). Again, slow relaxation of the contact potential (surface photovoltage) was observed.

Logarithmic time dependence of conductivity has been recognized as finger-print of Coulomb Glass behavior in disordered electron systems. Coulomb glasses are systems with states localized by disorder and long-range interactions between their particles. Up to now Coulomb Glass behavior has been found mostly at deep temperatures in granular metals. Interestingly, even though conducting polymers have the main ingredients needed for Coulomb Glass behavior –disorder and localized states- and although the conductivity in these systems is not yet well understood, this phenomenon has not been studied in this field. On the basis of our experimental results we propose that Coulomb Glass behavior may be relevant in conducting polymers. Indeed, our studies indicate that the electrostatic properties of the polymers investigated are closer to a glass-like material than to typical insulating, semiconducting or metallic matter.

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Figure

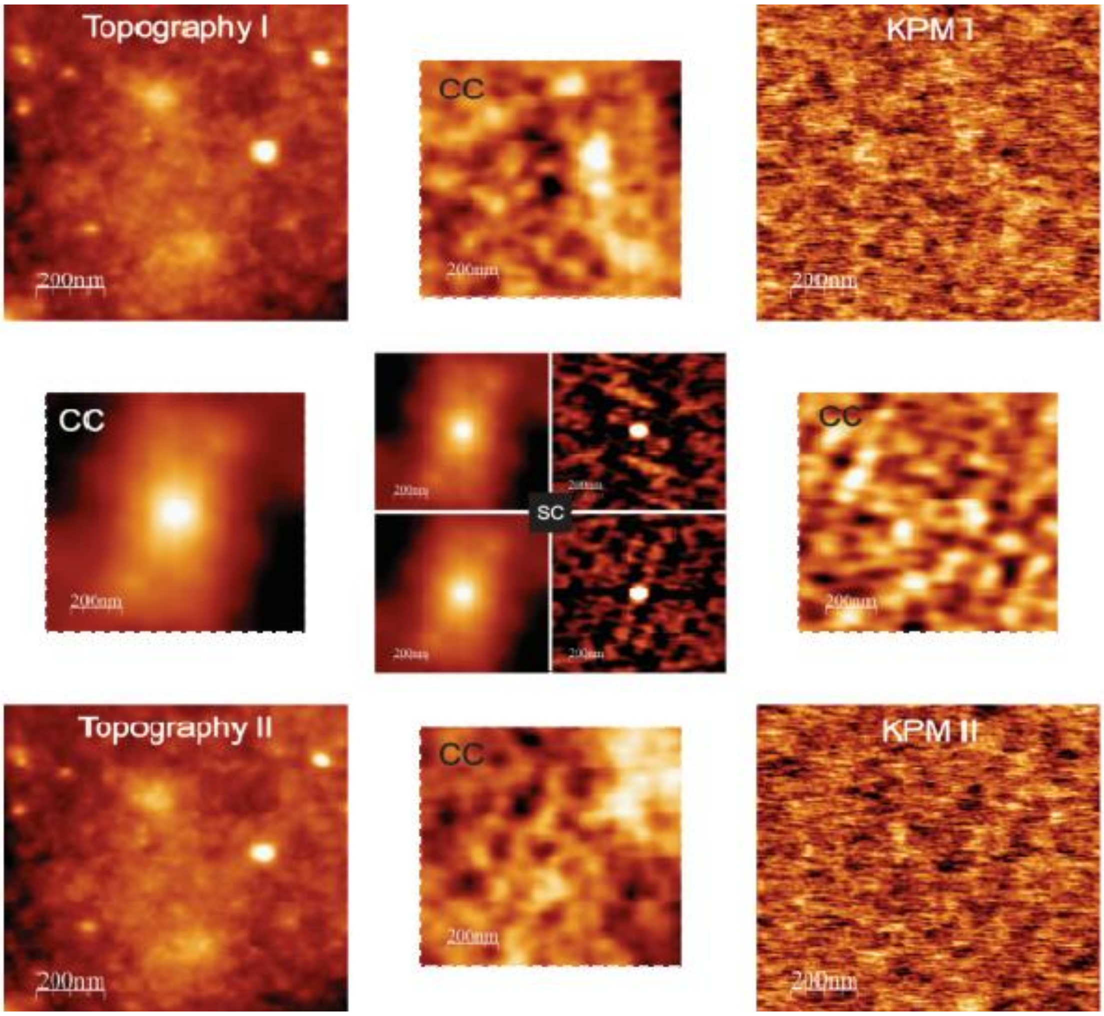

Figure1: Images showing the topography (top and bottom left) as well as the measured Contact Potential (top and bottom right) of a Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] sample. The upper experimental images (Topography 1 and KMP1) were acquired shortly (a few minutes) before the lower images (Topography 2 and KMP2). The images marked CC show the cross correlation between the corresponding experimental images. We find strong correlation (centre left image) between the topographic images, no correlation (top and bottom images in the middle) between simultaneous images of topography and contact potential and no correlation (centre right) between successive Contact potential images. The image in the centre shows the self-correlation calculated for each of the four experimental images

Controlling graphene plasmons with resonant metal antennas and spatial conductivity patterns

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In general, noise is a nuisance that limits the quality of experimental measurements. From a fundamental point of view, generally either statistical or quantum noise limit the ultimate resolution. On the other hand, according to the dissipation-fluctuation theorem, (some) noise is related to the dissipation of a physical system. In the present work, we will discuss noise in the tip-sample interaction of a Scanning Force Microscope (SFM), and investigate what information can be extracted from this noise. We will show, on the one hand, that thermal noise allows to precisely calibrate the oscillation amplitude in Dynamic SFM [1,2]; and on the other hand that it is possible to acquire "noise" images showing a well defined pattern, different from topography or other acquisition channels.

A typical electronics used to process the dynamic motion of the cantilever can be adjusted to transfer the thermal noise of the cantilever motion from its resonance frequency to a much lower frequency within the typical bandwidth of the corresponding electronics. If the force constant of the cantilever is known then the oscillation amplitude can be precisely calibrated from the thermal power measured in the output signals of a lock-in type dynamic SFM electronics.

For typical applications thermal noise is considered the most important noise source in a SFM setup. We show, however, that additional noise sources may be present in Dynamic SFM measurements, which may be significantly larger than the well-known thermal noise. Using different SFM techniques (Force Spectroscopy, Kelvin Probe Microscopy, 3D modes, etc), we analyze this noise for amphiphilic molecules (SDS) adsorbed on a graphite substrate. Our experiments show that the different chemical nature of these materials induce a different magnitude of the measured noise, leading to an image with "chemical" contrast (see figure 1).

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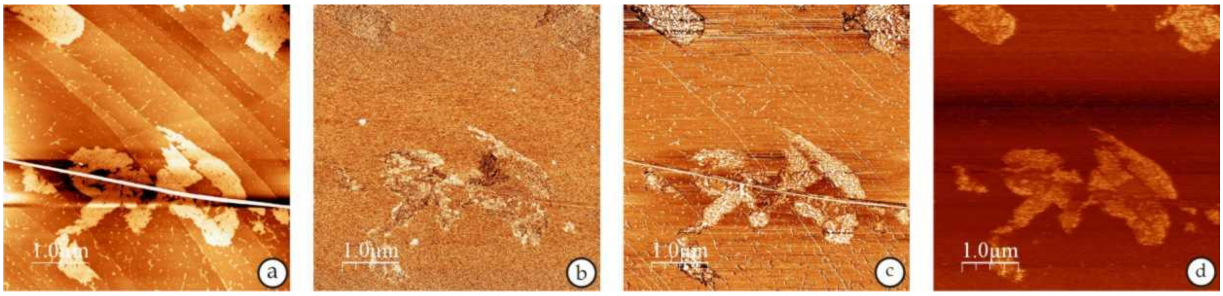

Figure1: Topography (a), frequency shift (b), contact potential (c) and noise images (d) acquired simultaneously in the so called Amplitude Modulation mode (topography feedback performed keeping the oscillation amplitude constant).

Tracing the formation of a non-covalent bond between two neutral molecules

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Chemical forces and conductance, during formation of covalent bonds in atomically sharp metallic junctions, depend exponentially on the leads distance. At the transition between tunneling and point contact short range interactions induce structural relaxations, the electronic structure gets locally modified and ultimately the tunneling barrier collapses [1].

Here we explore the extreme case of interaction between two neutral (closed shell) molecules: a carbon monoxide (CO) placed on an STM tip and an acetylene molecule (C_2H_2) adsorbed on a Cu(111) surface. The molecules are forced to interact when the CO is brought into the C_2H_2 close proximity.

By means of combined STM and nc-AFM measurements and DFT and transport simulations we characterize with unprecedent resolution the formation of a weak intermolecular electrostatic interaction by careful analysis of short range forces, conductance and their spatial variation (Fig. 1).

We find that the attractive force acting between the CO-tip and the acetylene molecule consists on London dispersion and local electrostatic forces arising from the charge redistribution induced by the proximity of the two molecules (Fig. 1b). No trace of charge accumulation is found in the gap between the two molecules which rules out the formation of a covalent bond.

This passive intermolecular coupling protects the tunneling barrier from collapsing even in the repulsive force regime, stabilizes the contact and allows quantifying subtle conductance and force variations along the small C_2H_2 molecule.

Moreover when the molecular junction is compressed for sufficiently large repulsive forces the CO molecule bends at the tip, the stiffness of the junction decreases and new channels of electron transport are open.

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Figures

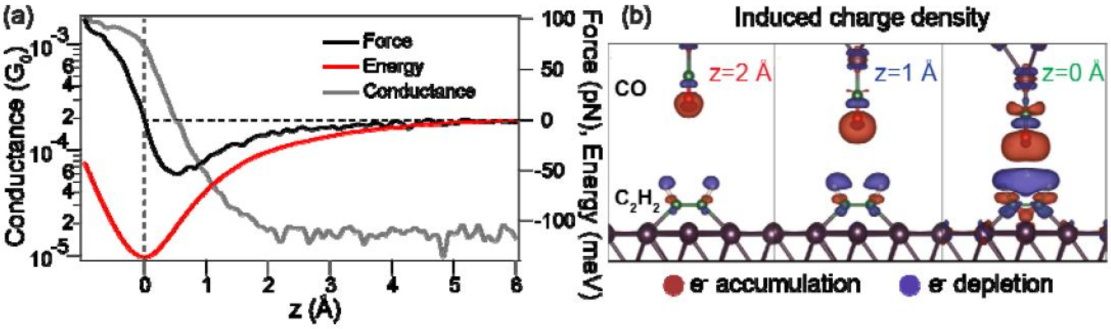

Figure1: (a) Conductance, force and energy curves taken with a CO functionalized tip at the C=C bond on a C_2H_2 molecule. A change in conductance occurs at the energy minimum, on the contrary to covalent bonds where it happens at the force minimum [1]. (b) Plot of the induced electron charge density projected in 1D upon CO tip approach.

Microwave Impedance Microscopy: Visualizing Electrical Property in Complex Quantum Materials

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Advances in the study of electronic materials have been one of the driving forces for both scientific and technological developments in the recent decades, from transistors to quantum devices, from superconductors to topological insulators. Various SPM techniques have been developed as both the result and the cause of our ever-increasing understanding and ability to build and control complex material properties and device structures. In this tutorial, I would like to introduce Microwave Impedance Microscopy (MIM), a unique SPM technique that is capable of measuring electrical property at the nano-scale.

MIM measures the microwave response of the impedance formed at the tip-sample interface. Working in the microwave frequency range (1-10 GHz), it balances the capacitive coupling and sample resistive response, which enables characterizing conductivity ranges from metals to insulators. In particular, this frequency is low enough that only low energy excitations are involved in the response, while it is also high enough that no counter-electrode is necessary on the sample, therefore minimizing the sample preparation. Plus, it also has the ability to probe sub-surface features. Over the years, we have built advanced instrumentation and developed well-shielded cantilever probe that can efficiently deliver microwave signals, which allow achieving spatial resolution below 50 nm and electrical sensitivity approaching zF (10-18 F).

I will present examples of applying MIM to study complex quantum materials. In colossal magnetoresistance materials, we visualize the percolative nature of the metal-insulator transition and the associated glassy dynamics. In ferroelectric materials, we investigate the conductive domain wall structure. In topological materials, we study the edge state configuration in quantum Hall effect and quantum spin Hall effect. These case studies demonstrate the capability of the MIM technique and the new insights it provides on the understanding of physics in complex quantum systems.

Real Space Imaging of Quantum Hall Edge States in Graphene with Microwave Impedance Microscopy

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In the presence of a magnetic field, a two-dimensional electron gas enters into the quantum Hall state associated with formation of the Landau levels. At exact filling of Landau levels, carriers in the bulk become localized while only the edge states serve as conduction channels responsible for the quantized transport resistances. Spatial reconstructions of these edge states can occur for a non-uniform carrier density profile due to electrostatics, giving rise to incompressible regions that separate the edge states from the bulk. In this work we use microwave impedance microscope, a newly developed near-field scanning probe microscope, to image the quantum Hall edge states in graphene devices. Scanning images clearly show dividing regions of insulating bulk and conductive edges. We study the evolution of the edge patterns as the carrier density is tuned through multiple Landau levels. In particular, a finite voltage bias on the tip can induce a local charge perturbation, which leads to an extra incompressible ring that moves along with the tip during scanning. Such incompressible ring can be used to probe the variations of the local carrier profile. Our results indicate that the carrier density in graphene tuned by the back gate tends to increase toward the edge due to electric screening. This is in contrast to the case of conventional semiconductor two-dimensional electron gas systems, where the carrier density always decreases toward the edge due to charge depletion. We will discuss how this charge profile affects the formation of the incompressible stripes.

Revealing reaction mechanisms by direct visualization of covalent bond structure of reactants, intermediates and products.

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Observing the intricate chemical transformation of an individual molecule as it undergoes a complex reaction is a long-standing challenge in molecular imaging. Advances in scanning probe microscopy now provide the tools to visualize not only the frontier orbitals of chemical reaction partners and products by scanning tunneling microscopy (STM), but their internal covalent bond configurations as well, by means of non-contact atomic force microscopy (nc-AFM).[1,2] We have used nc-AFM to investigate reaction-induced changes in the detailed internal bond structure of individual oligo-(phenylene-1,2-ethynylenes) on various metal surfaces as they underwent a series of cyclization processes (Fig. 1).[3] Our bond-resolved single-molecule imaging, together with DFT calculations, allows us to extract an exhaustive picture and unparalleled insight into the chemistry involved in complex enediyne cyclization cascades.

The detailed mechanistic understanding can in turn guide the design of precursors for the rational synthesis of functional molecular architectures by means of surface-supported chemistry.[4] However, good care must be taken of the substrate because its catalytic influence can drive the reactions of the same reactants in disparate directions. Moreover, substrates may act as a heat-sink for the energy released in exothermic reactions, explaining the experimentally observed metastability of certain intermediate species (Fig. 2).[5] The surface selectivity towards particular intermediates in the dissipation process could in fact open up new venues for a tailored stabilization of radical species, with great potential impact in heterogeneous catalysis.r.

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Figure

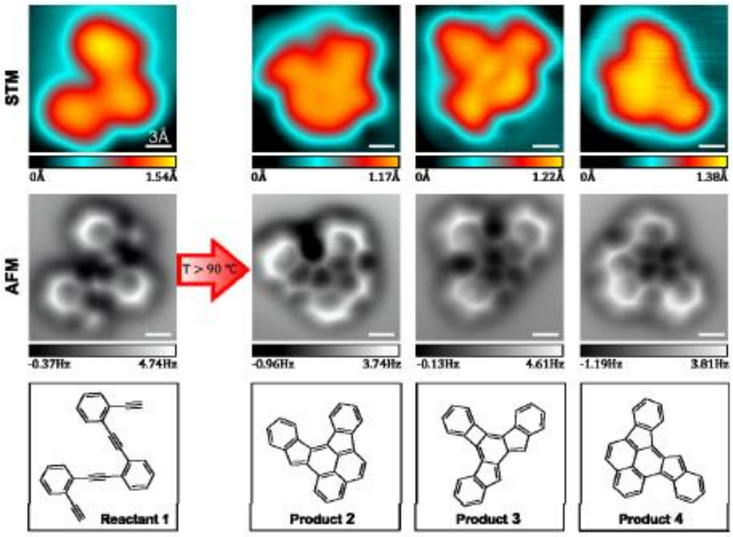

Figure1: Scanning tunneling microscopy (top row), nc-AFM images (middle row) and wireframe molecular structures of the reactant and the three most common products resulting after annealing the Ag(100) sample to temperatures above 90 °C.

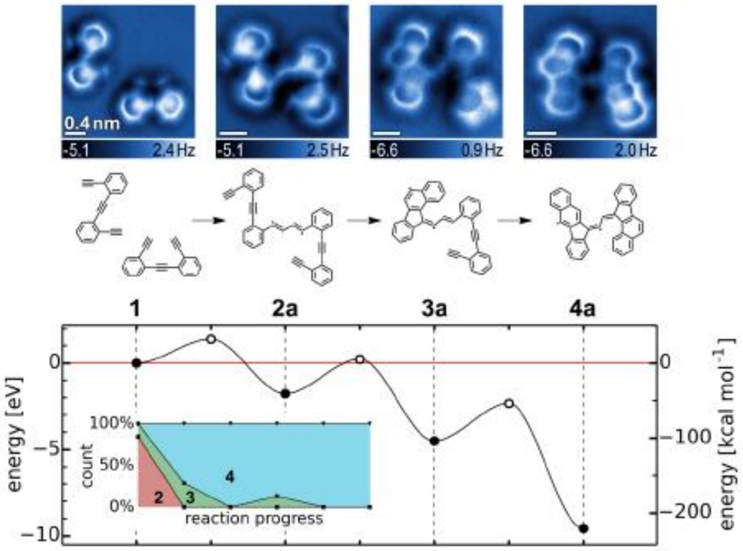

Figure2: Nc-AFM images (and derived wireframe structures) show the stepwise transformation on Ag(100) of 1 via 2a and 3a to 4a. DFTB calculations of the energy landscape show that the energies of these species (filled circles), as well as the energies of the transition states (empty circles) decrease with each step. The shift of the relative abundance of the species within this reaction pathway (2a, 3a, 4a) with each subsequent annealing step is plotted in the inset.

Optically induced forces in scanning probe microscopy

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As technology relies more and more on the design and engineering of structures which are nanoscale in two or three dimensions, it becomes increasingly important to have metrology tools capable of characterizing the properties of the constituent materials at the relevant length scales. In the field of nano-photonics, an additional requirement is to measure properties of electromagnetic fields at scales much smaller that the wavelength. Such demands are usually met by scanning probe microscopy (SPM) which, in general, relies on detecting the consequences of the field-mediated interaction between a small probe and some physical surface.

A myriad of microscopy techniques have been developed to map fields characteristics by collecting radiation that is either coupled through or scattered off a subwavelength probe. What makes optical measurements extremely challenging however is the fact that detecting radiation typically involves some sort of photon-to-electron conversion. Moreover, guiding or scattering light at subwavelength scales usually involves interactions with metallic structures leading to inevitable thermal effects.

We will review the phenomenology of a near-field light measurement technique based on the detection of optically induced forces acting on scanning probes . This type of detection overcomes some of the above limitations and permits true broad-band detection of light directly in the near-field. Without sacrificing the spatial resolution, this measurement modality not only circumvents the need for photon detectors, especially interesting for IR and THz, but it also complements traditional approaches for mapping the 3D properties of electromagnetic fields . Because the magnitude of field-probe interaction can be conveniently controlled, the optically induced forces can also assist in reconstructing other properties such as the profiles of weak surface forces of different origins.

The physical background and the main characteristics of optical force detection will be reviewed together with practical applications of interest for the broader field of the scanning probe microscopy.

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Spin Hall Effect of Light and Some Mechanical Consequences

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Electromagnetic waves carry angular momenta. For waves in pure states of polarization, the spin-orbit interaction results in a spiraling power flow, which is determined by the extent of wave-matter interaction. The phenomenology is similar to the electronic spin Hall effect and it provides essential insights for developing new sensing approaches at nanoscales. In addition, the laws of momenta conservation lead to remarkable mechanical effects as a result of light interacting with spherically symmetric, optically isotropic, and lossless objects. Conservative and nonconservative optical torques can be generated and, most importantly, their interplay can be controlled by the polarization of light. This action induced by optical crosstalk creates a new kind of "optical matter" with mechanical properties strongly coupled to the electromagnetic field.

Trapping of gold adatoms by dimethyl sulfoxide on Au(111)

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Dimethyl sulfoxide (DMSO) is a polar, aprotic solvent that is commonly used both in laboratory and industrial processes. Despite its widespread use, the interaction of DMSO with metal surfaces has been only marginally studied, and the atomic scale details of this interaction are yet poorly known. In this work we characterized the adsorption of DMSO on the (111) surface of gold by means of low temperature scanning tunneling microscopy (LT-STM). The STM images revealed the tendency of DMSO molecules to form complexes made by up to four molecules and structural models for each complex were proposed. By comparison of experimental and simulated STM images, based on density functional theory (DFT) calculations, we were able to demonstrate that some of these complexes have the peculiar characteristic of entrapping inside them one or two gold adatoms. Albeit being invisible in the STM images, DFT calculations reveal unambiguously that the presence of these adatoms is crucial for the stability of the complexes. These findings can be of relevance to gain more insight into the behavior of DMSO in electrochemical processes, where the interaction of DMSO with metal electrodes is crucial, or when exploiting DMSO as a solvent for the surface deposition of molecules by electrospray ionization or drop casting.

STM probing of spin excitations in nanoengineered spin chains

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Scanning tunneling microscopes (STM) can be used to build and probe chains of a few magnetic atoms on a surface, opening the venue for atomic scale engineering of nanomagnets. STM Inelastic spectroscopy permits to probe the spin excitations of these designer nanomagnets. Combined with the atomic resolution of the STM, this affords the imaging of the spin excitations atom by atom. This concept will be illustrated in the context of recent experiments [1] where chains with 5 Fe atoms deposited on a Cu2N/Cu surface are coupled ferromagnetically and their spin waves are mapped with atomic scale resolution. The use of this technique to map exotic fractional edge states in S=1 spin chains will also be discussed.

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Magnetic Imaging and Manipulation of Molecular-based Nanoparticles

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In the race towards the miniaturization in nanoelectronics, magnetic nanoparticles (MNPs) have emerged as potential candidates for their integration in ultra-high density recording media. Molecularbased materials open the possibility to design new tailor-made MNPs with variable composition and sizes, which are benefited by the intrinsic properties of these materials. Among them, bimetallic, cyanide-bridged nanoparticles of the family of Prussian blue analogues (PBA-NPs) have been broadly studied.[1] Before their implementation in real devices is reached, a precise organization on surfaces and a reliable characterization and manipulation of their individual magnetic behavior are required. We will show how molecular-based MNPs are accurately organized on surfaces by soft-lithography and how their individual magnetic properties are detected and tuned by means of low-temperature magnetic force microscopy[2] (LT-MFM) with variable magnetic field. The magnetization reversal on dispersed and organized MNPs is investigated, and the temperature dependence of their magnetic response is evaluated

Finally we will present the results from magnetic measurements performed with a functionalized-MFM tip by a PBA-NP (Figure 1). The new contrast observed with this PBA-NP-tip is compatible with an inplane magnetized MFM tip (as opposed to the original out-of-plane MFM tip). Therefore, this functionalization converts the ~50 nm tip into a ~19 nm tip with an effective magnetic moment in the +x direction, enriching the information that can be extracted from the magnetic images.

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Figure

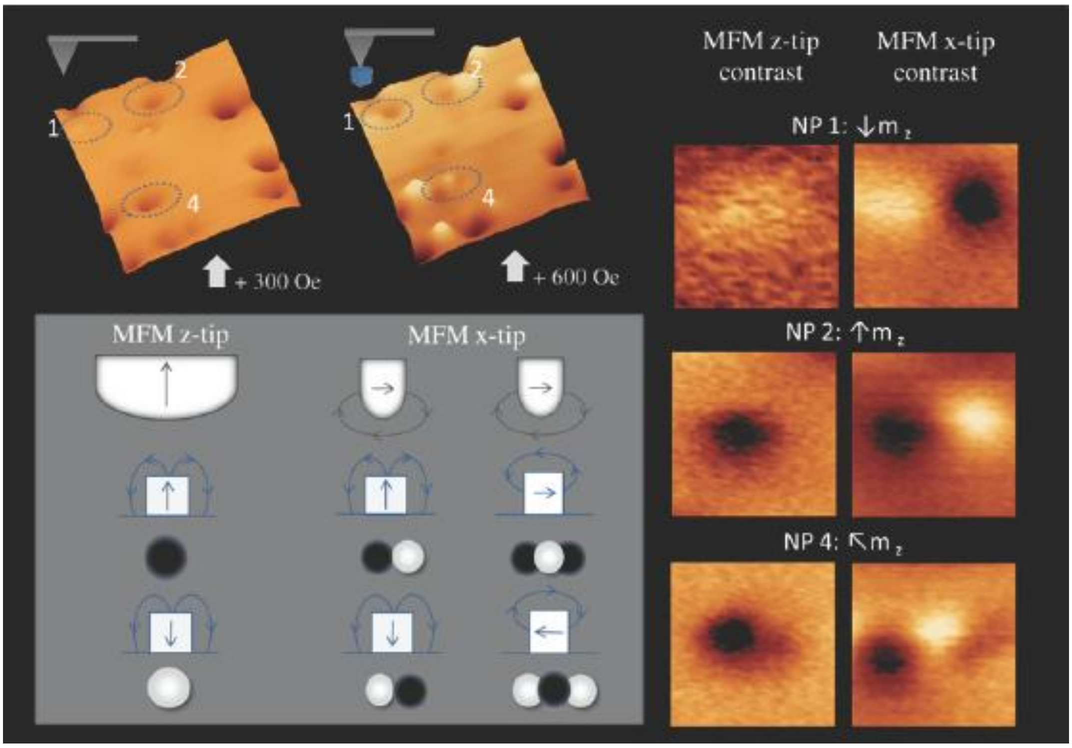

Figure1: 3D magnetic images (520 nm x 520 nm) acquired with the regular MFM tip (left) and the functionalized PBA-NP-tip (right). Three NPs are highlighted and zooms in of 115 nm x 115 nm show the different magnetic contrasts for each case. A schematic of the magnetic contrast for the different situations is shown for clarity.

The atomic force microscope and its relation to the tunneling microscope

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After Gerd Binnig and Heinrich Rohrer invented the scanning tunneling microscope, a variety of other scanning probe microscopes appeared and the atomic force microscope became the most widely applied child of the scanning tunneling microscope. The talk covers the early years of atomic force microscopy with it's ties to the STM as well as very recent developments where STM and AFM are reunited.
Some personal memories with fruitful interactions with Heini Rohrer are included

Electric AFM modes: Current sensing AFM and Electrostatic Force Microscopy

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In this lecture the basis and principles of the main electric AFM modes, namely, current sensing AFM and electrostatic force microscopy, will be presented. Electric AFM modes are characterized, in general, by the use of conducting AFM probes and by the application of an electric voltage between the probe and the sample with the objective to determine a local electrical property, in addition to topography. In current sensing AFM one mesaures the local electric current flowing through the sample in response to an applied voltage, from which information on the local electric conduction properties can be obtained. If the applied voltage is time varying (ac voltage) then one can measure the local electric impedance, which in this case can provide information on both the local (ac) conductance and local capacitance, from which information on the local conductivity and dielectric constant of the sample can be obtained. On the other hand, in electrostatic force microscopy one measures the electrostatic force acting on the probe in response to the applied voltage. From the measured electric force one can obtain information on the local charge or potential of the sample, as well as, on its local dielectric properties. Examples of applications of the different techniques will be provided, togther with a short discussion on its quantitative interpretation.

Electric polarization properties of single nanoparticles and viruses measured with electrostatic force microscopy

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I will describe the use of electrostatic force microscopy to determine in a quantitative manner the electric polarization properties (i.e. the dielectric constant) of small scale systems such as single nanoparticles and single viruses. The method consists in measuring the electrostatic interaction force between a biased sharp conducting tip mounted on a force sensing cantilever and the sample under study, and then using specific calibration procedures and finite element numerical calculations to quantify the force. The method is demonstrated by its application to 10 nm radii polystere, silicon dioxide and silicon nitride nanoparticles, as well as to single T7 bacteriophages, and its corresponding empty capsids. I will discuss the possibility to use this technique as a novel label free nanoscale composition sensitive technique.

Recovery of Permittivity and Depth from Near-Field Data as a Step toward Infrared Nanotomography

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Scattering-type scanning near-field optical microscopy (s-SNOM) is a powerful optical technique for nondestructive spectroscopic imaging with deep subwavelength resolution [1]. In s-SNOM, images are recorded by scanning a sharp illuminated probe along the sample surface and detecting the backscattering. This scattering depends on the near-field interaction between the probe and the sample, thus revealing the dielectric properties of the sample in the image contrast. While s-SNOM has demonstrated the ability of quantitative analysis of such images for samples with structure varying in two dimensions [2], the elucidation of sample structure in all three dimensions has proved elusive due to complicated nature of near-field interactions.

In this work we open the door to the third dimension for near-field techniques by introducing and providing the first experimental demonstration of a method for performing a rapid recovery of the thickness and permittivity of simple 3D structures, such as thin films and nanostructures made of weak oscillators [3]. This is accomplished via a novel nonlinear invertible model of the near-field scattering, taking advantage of the near-field data recorded at multiple harmonics of the oscillation frequency of the near-field probe as depicted in the Fig. 1.

Our work enables the quantitative nondestructive nanoscale-resolved optical studies of thin films, coatings and functionalization layers, as well as the structural analysis of multiphase materials and other samples in which the topography does not correlate with the chemical or optical properties. It opens new frontiers for chemometrics, materials and bio sciences and represents a major step towards the further goal of a general tomographic reconstruction of the sample.

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Figure

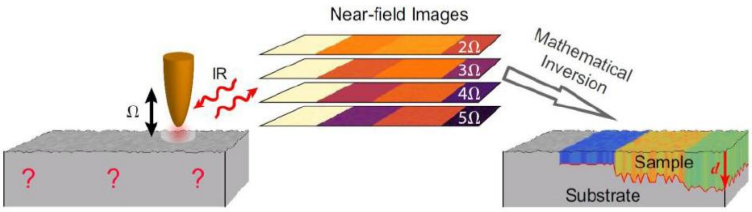

Figure 1: The samples structure in three-dimensions (here the permittivity and thickness) can be recovered from near-field images recorded at multiple harmonics of the tip oscillation frequency.

STM+IETS: measuring low energy excitations at the atomic scale

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Scanning tunneling microscopy (STM) is a powerful tool for probing the structural, electronic, and magnetic properties of metallic, semiconducting, superconducting, and even thin-insulating surfaces. The exponential dependence of quantum mechanical tunneling through a potential barrier makes the current measured by STM sensitive to changes in the distance between the tip apex and underlying sample at well below atomic dimensions, enabling atomically resolved imaging on the surfaces of many materials. Often, however, variations in density of states (DOS) also occur across the surface; these also modify the total tunnel current and therefore give rise to apparent topographic contrast. This makes interpretation of STM images more difficult but can also be exploited to yield one of the STM most powerful capabilities: spatially resolved spectroscopy. Using changes in the current as a function of applied bias voltage across a tunnel junction to measure DOS – often referred to as elastic tunneling spectroscopy – was first performed in measurements of the superconducting gap in planar tunnel junctions [1]. Combined with the spatial resolution of STM, elastic tunneling spectroscopy – often called scanning tunneling spectroscopy (STS) – allows for the mapping of the local DOS (LDOS) of surfaces at the atomic scale. This has revealed a variety of phenomena, ranging from standing waves of electronic surface states [2] in metals to novel ordering in high-Tc superconductors [3].

While accessing the spatially resolved LDOS of a surface can reveal substantial information about its electronic and magnetic properties, additional insight can also be obtained by probing the low energy excitations in the system. This was also accomplished many years ago in planar tunnel junctions using inelastic electron tunneling spectroscopy (IETS), and was first used to reveal the vibrational modes of molecules trapped in the tunnel junction [4]. In a typical IETS process, there is a sharp change – usually an increase but sometimes a decrease – in the tunneling current above a certain voltage that corresponds to the energy needed to create an excitation in the tunnel junction. Measuring the voltage of this onset therefore directly yields the energy needed to create the excitation, making the process analogous to other inelastic scattering techniques like Raman scattering or Electron Energy Loss Spectroscopy (EELS). When combined with STM, however, IETS can be used to measure the low energy excitations of individual atoms and molecules on surfaces, a sensitivity that most other inelastic scattering techniques cannot match.

In this tutorial we will cover the basics of inelastic tunneling spectroscopy. After introducing the early tunneling spectroscopy experiments in planar junctions, we will focus on STM-based measurements. Specifically, we will begin by reviewing the first STM-IETS experiments, which were used to measure the vibrational modes of individual molecules on surfaces [5]. From this remarkable initial demonstration, we will move on to more recent developments in which the rotational [6,7] and spin [8] degrees of freedom in single atoms and molecules can be excited.

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Using Electronic Coupling to Control Magnetic Properties at the Atomic Scale

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The drive to continue Moore's Law by shrinking electrical components down to the ultimate limit has led to a great deal of interest in atomic and molecular-scale electronics, in which individual atoms and molecules can be used as circuit elements. More recent proposals also seek to exploit the magnetic properties of these nanoscale objects in new applications in information technology and spintronics. In typical device geometries, the magnetic element is coupled to electrical leads, and these interactions can strongly affect the properties of the quantum system.

Using scanning tunneling microscopy and spectroscopy, we study the effects of interactions between individual magnetic atoms and molecules that are separated from an underlying metallic surface by a thin-insulating layer of copper nitride (Cu2N). For Co atoms on large Cu2N islands, we find that exchange coupling of the spin to the metallic bath can result in Kondo screening as well as dramatically shifting the energy levels of the spin and modifying its effective magnetic anisotropy [1], the property that determines the stability of its spin orientation. By controlling the exchange coupling, we can tune both the Kondo screening of the systems as well as the anisotropy energy over a broad range of values. Furthermore, this system constitutes one of the few cases in which an open quantum system's energy levels, rather than just its excited-state lifetimes, can be controllably and observably renormalized.

* This work was done in collaboration with Jenny C. Oberg, M. Reyes Calvo, Fernando Delgado, María Moro-Lagares, David Serrate, David Jacob, and Joaquín Fernández-Rossier.

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Probing the local work function with tunneling current decay constant measurements

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Performing AFM with a conducting tip allows the local work function of the surface to be investigated. The most common technique is Kelvin Probe Force Microscopy. However, with this technique, simultaneous STM is difficult, because of the varying bias potential. We propose an alternate technique [1], first introduced by the STM community [2], in which measurements of the tunneling current at the oscillating frequency of the tip can yield information about the vertical decay constant of the tunneling current, κ. This decay constant is related to the local work function and the electronic states being probed. Our technique can be performed at room temperature in combination with STM and AFM.

We show atomic-scale measurements of both a bare Si(111)-7×7 surface and a Ag/Si(111) surface. We discuss the complications in converting from a recorded κ value to the local work function [3].

Furthermore, we study PTCDA islands in which neighboring molecules have different electronic structures. These differences are hard to detect in normal AFM data [4], however are clear to see in the κ channel (see Fig.).

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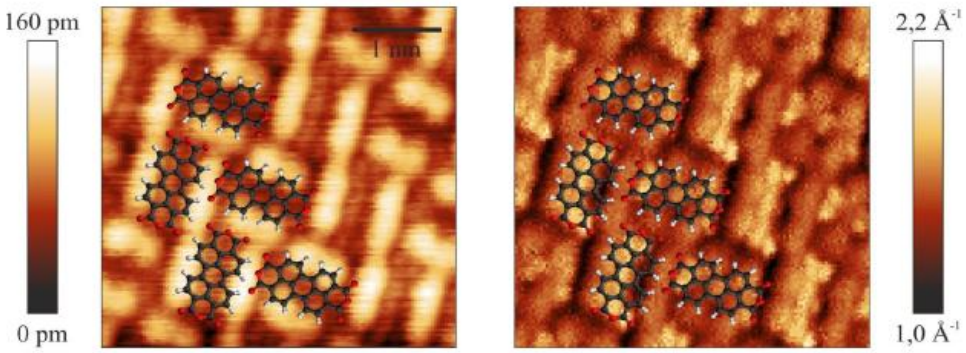

Figure1: topography (left) and κ channel (right) of a PTCDA island.

STM and Gating Nanopores for Single Molecule DNA and RNA Electrical Sequencing

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STM and Gating nanopores are the key Nano-Biodevices for the third generation DNA sequencing technologies. These nanodevices will make sequencing kilobase length single-stranded genomic DNA and RNA, or identifying individual small molecules using only electric currents and without fluorescent labels at low cost and unheard speeds.

Nanopores approximately 2-30 nm in diameter have been formed on a Si substrate, and nanogap electrodes with spacing equal to the diameter of the DNA molecules are fabricated in the Si_3N_4 membrane (see Fig.). This nanostructure is expected to detect molecules passing through the nanopore by changes in the electric current flowing between the nanogap electrodes. The electric current passing between the nanoelectrodes comes from a tunneling current conducted via molecules.

We have demonstrated identifying single-nucleotides using these nano-fabricated devices by measuring tunneling current across the nanogap electrodes. And, we found that the single peak current on current histograms determines the single-molecule conductivity of the order of dGMP >dAMP >dCMP >dTMP, and rGMP > rAMP > rCMP > rUMP. Methylated dCMP and oxo-dGMP which play important roles in epigenomics are also distinguishable. Recently, we have succeeded in sequencing DNA Codons and Micro-RNA by assembling contigs using a pair of nanogap electrodes. For micro-RNA, Let7 families which are recognized as the biomarkers for cancer have been successfully identified by this method. These results provide an essential scientific basis toward constructing Innovative Nano-Biodevices for the emerging DNA, RNA and Protein sequencing technology.

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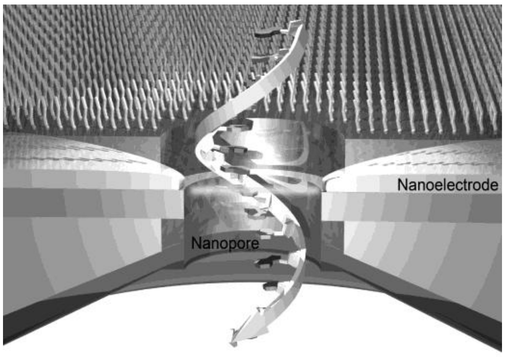

Fig. Schematic diagram of gating nanopore devices.

Nanoscopy by scattering infrared light off a metal probe tip

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This contribution reviews history and basic principles of the infrared near-field microscope (of scattering type, s-SNOM). Its starting point was a suggestion by Synge in 1929. Its experimental realization (first by Wickramasinghe in 1990) apparently had to await the invention of scanning-probe nanoscopies, primarily the STM by Binnig and Rohrer in 1981. Initially surprising, the interaction by light of a tip with a sample under test can be as highly confined spatially as is the interaction by current or force. Thus, a scanning-probe nanoscope can simultaneously produce topography and optical maps with spatial resolution better than 20 nm. The use of broadband-infrared illumination allows since recent the determination of the local chemical composition, structural identity, and electrical conductivity.

Infrared nanoscopy of biological and extraterrestric minerals

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In scattering-type scanning near-field microscopy (s-SNOM) the strongest response is observed in the mid-infrared for two reasons, (i) the antenna function of standard metallized tips being optimal in the mid-IR, decreasing significantly towards the near-IR, and (ii), the mid-infrared occurrence of characteristic lattice vibrations (optic phonons) of crystalline sample materials. This is fortunate since many interesting materials contain rich nano-composite crystalline structures. Recent results of infrared nanoscopy will be discussed for nanocrystals found in mussel shells and human bone, and in comets and meteorites.

Physical concepts of scanning tunneling microscopy and its application to excitations and magnetic substrates

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In this tutorial I will briefly review electronic currents on the nanoscale, the different physical concepts associated with electronic transport and their application to the scanning tunneling microscope (STM). I will next explore the customary approximations to understand STM images, its application to magnetic substrates using spin-polarized tips and the meaning of the different aspects of constant current images. I will also explore the possibility of producing excitations with the tunneling current and how this has been used to develop a very local spectroscopy that can give the signature of vibrations or magnetic excitations of a few atoms.

Spontaneous magnetic switching due to spin decoherence

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Our calculations permit us to understand the thermal and non-thermal spin switching found by Loth et al. [Science 335, 196 (2012)] when studying antiferromagnetic chains of Fe atoms. Decoherence fixes a given spin ordering by quenching the Rabi oscillations between equally probable configurations. This decoherence is induced by the substrate electrons and has a characteristic 1/T dependence with temperature. Our calculations show that quantum tunneling is not at the origin of the experimental low-temperature behavior unless unrealistic low temperatures are attained.

IETS+Time: resolving spin dynamics between milliseconds and picoseconds

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Perturbative probes are essential for the exploration of magnetism. Traditionally external magnetic fields are used to perturb the magnet's ground state configuration. Here I will discuss how inelastic electron tunneling spectroscopy and electronic pump probe spectroscopy can be used in a scanning tunneling microscope to characterize the spin dynamics of few-atom nanomagnets on surfaces.

Inelastic electron tunneling appears as steps in the differential conductivity. The voltages at which these steps occur directly relate to the energy of a spin excitation in the atom, molecule or surface under investigation. To obtain information on the fast dynamics of the spin we make use of another property of inelastic tunneling: after every inelastic tunneling event the spin is in an excited state.

I will introduce two methods that capitalize on the modified interaction of tunneling electrons with the excited spin. Firstly, spin pumping (Nature Phys. 6 340, 2010) in which we record IETS spectra with increasing tunnel junction conductance and model observed changes in the spectral shape using master equations to extract the characteristic spin relaxation time of every spin state involved in the inelastic tunneling. Secondly, all-electronic pump probe spectroscopy (Science 329 1628, 2010) in which we use a train of fast voltage pulses to repeatedly excite magnetic atoms out of thermal equilibrium and monitor the subsequent dynamical evolution by time-delayed weaker voltage pulses. In contrast to spin pumping, pump-probe spectroscopy operates directly in the time domain and proves extremely sensitive to minute variations in the spin relaxation time.

Spin interaction in atomically assembled quantum magnets

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Magnetic atoms on surfaces can interact with each other if they are placed into close proximity. On monolayer thin insulating films the magnetic atoms retain their quantum character and are well-described as spins with quantized states. We use the probe tip of a low-temperature scanning tunneling microscope to arrange transition metal atoms into arrays of our own design and create experimental representations of different Spin Hamiltonians.

The spin magnitude of each site can be set by choosing different transition metal elements and spin-spin interaction can be adjusted by different geometric arrangements. Inelastic electron tunneling and all-electronic pump-probe spectroscopy at GHz frequencies quantifies the energy level structure, energy loss mechanisms and spin lifetimes of the interacting spins (Science 329 1628, 2010). Using this technique we identified a new approach to suppress magnetic tunneling in antiferromagnetic spin chains (Science 335 196, 2012), observed emergent many-body states akin to the Kondo effect and found a mechanism to tune spin states controllably by exchange interaction from the STM tip.

The combination of atom-by-atom assembly and time-resolved inelastic electron tunneling spectroscopy provides a new toolset to explore fundamental quantum mechanics in artificial correlated electron systems.

Probing the magnetic interaction between single Cr atoms

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In the last decade it has been shown that magnetic properties of a single atom can be probed by inelastic electron tunneling spectroscopy [1]. In addition, magnetic nanostructures have been routinely assembled via atomic manipulation [2]. In the tunneling junction, spin excitation is induced if the bias voltage exceeds the threshold energy for excitation. The spin excitation process allows the electrons to tunnel inelastically leading to a stepwise increase in the conductance over the threshold bias [1,2].

Here we aimed to study the magnetic characteristics of isolated Cr atoms adsorbed on Cu2N/Cu(100) surface. Cr atoms were deposited and their properties were investigated at 1.1 K in a SPECS JT STM that has a magnetic field up to 3 T perpendicular to the sample.

Different inelastic tunneling spectra were observed over Cr atoms adsorbed on N sites than on Cu sites. In the first case we interpret our spectra as interplay between spin excitation and Kondo screening effect, which appears here at larger biases. However, if the Cr atom adsorbed on the Cu site, the Kondo effect does not appear in the spectra, only a spin excitation has a contribution near the Fermi level. Our results suggest that the spin configuration of the Cr atom varies among different adsorption positions.

By atomic manipulation we have constructed Cr dimers with different interatomic spacing (see Fig). Among the investigated pairs, the strength of the coupling, J, was the strongest when the Cr atoms were occupying the nearest Cu sites separated by a single N atom (JA). The coupling becomes significantly weaker if the Cr atoms are placed diagonally (JB). Interestingly, increasing further the interatomic separation induces less significant quench in the coupling energy (JC). In a dimer, the close proximity of the Cr adatoms induces a local crystal strain. Beside the variation in the strength of the magnetic exchange interaction, the strain-enhanced anisotropy has a strong impact on the stability of the antiferromagnetic coupling [3]. The combination of both effects explains well our large variation of J.

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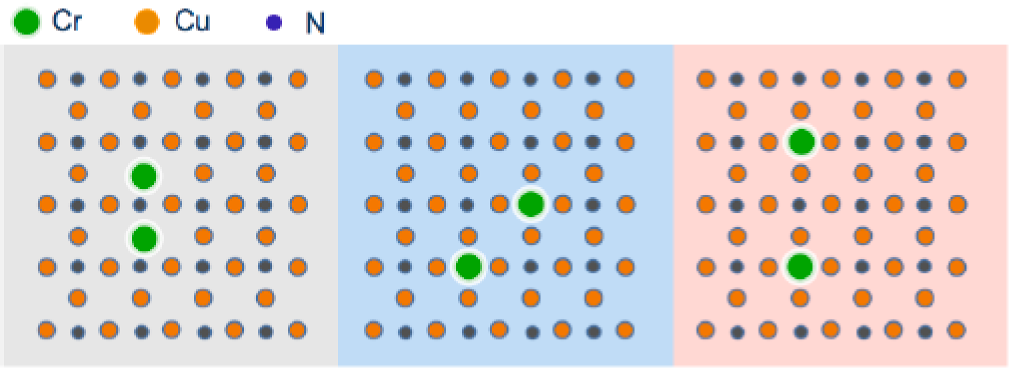

$$J_A(9.5\ \mathrm{meV}) \gg J_B(1.8\ \mathrm{meV}) > J_C(0.9\ \mathrm{meV})$$

$$d_A\,(3.6\text{Å}) < d_B(5\text{Å}) < d_C(7.2\text{Å})$$

Atomic Structure Affects the Directional Dependence of Friction

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Friction between two objects can be understood by the making, stretching, and breaking of thousands of atomic-scale asperities. We have probed single atoms in a nonisotropic surface (the H-terminated Si(100) surface) with a lateral force microscope operating in noncontact mode. We show that these forces are measurably different, depending upon the direction. Experimentally, these differences are observable in both the line profiles and the maximum stiffnesses. Density functional theory calculations show a concerted motion of the whole Si dimer during the tip-sample interaction. These results demonstrate that on an asperity-by-asperity basis, the surface atomic structure plays a strong role in the directional dependence of friction.

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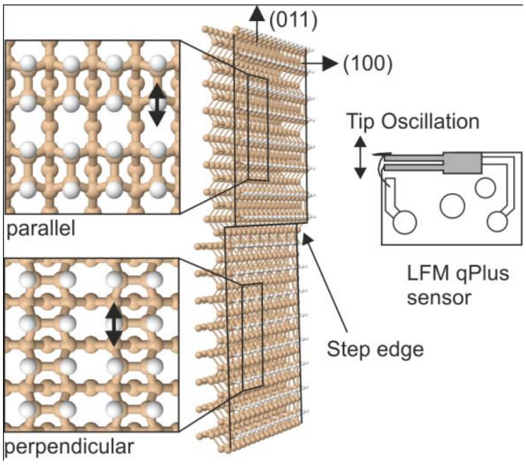

Figure1: Schematic of the experimental setup. By cutting the Si wafer on the {011} planes, the (011) crystallographic direction can be aligned with the tip oscillation. By moving from one terrace to another, data can be acquired with the tip oscillating either parallel or perpendicular to the Si dimmers.

High Resolution Dynamic Force Microscopy in a nutshell

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Frequency Modulation (FM) AFM, usually known as Non-contact AFM, has already proven its ability for imaging, chemical identification and manipulation at the atomic scale on both conducting and insulating materials. Semiconductor surfaces and UHV played a central role in the early development of the technique. FM-AFM has now matured and expanded into the study of challenging surfaces and biological material in liquids. In this talk, I'll review the basic principles behind the operation of FM-AFM, the theoretical developments necessary to describe the tip-sample interactions in technologically relevant materials like reducible oxides and carbon nanostructures, and the extra challenges involved in the imaging of surfaces and large biomolecules in their native liquid environment.

Reducible oxides such as CeO_2 and TiO_2 play a crucial role in the development of a green economy as catalysts for hydrogen production, efficient anodes for fuel cells and, in combination with organic molecules, as active elements in solar cells and novel transistor devices. The rich variety of contrasts observed and the unambiguous identification of the different chemical species, particularly in complex surfaces like anatase TiO_2(110), represent a challenge for FM-AFM. We'll show how the combination of force spectroscopy and theoretical calculations [1,2] provide a powerful tool to unravel the contrast mechanisms, to characterize the tip apex and to identify the nature of the visualized features. Further insight comes from tunneling currents, where their dependence on distance, bias voltage and tip apex can be exploited to achieve selective chemical imaging [3].

Carbon nanostructures, from fullerenes and nanotubes to graphene, their doped derivatives, and their interfaces with inorganic materials are expected to play a crucial role in the future development of electronic devices. A combination of FM-AFM with electronic transport measurements represents a really powerful tool to characterize simultaneously the structure and the electronic properties of these systems. We'll discuss in detail the interplay in the image contrast formation between the chemical bonding and the van der Waals (vdW) interaction in these materials [4], and the ability of FM-AFM to image the subtle charge modulations induced in graphene by its interaction with metals [5,6].

Finally, we'll introduce the challenges involved in simulating tip-sample interactions in liquids. We'll present some relevant technical issues, including the implicit versus explicit description of the solvent, the calculation of free energies and forces, and the use of steered molecular dynamics protocols. They will be illustrated with the study of large proteins like the IgG antibody (~13.000 atoms) adsorbed on hydrophobic surfaces like graphene [7].

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Graphene/metal Moirés: Can AFM resolve small electronic modulations?

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FM-AFM (usually known as NCAFM) has shown its ability to produce atomically-resolved images of all kinds of materials ranging from semiconductors to metals and oxides. Recently, NCAFM has been able to reveal the chemical bonds within polycyclic aromatic molecules and to characterize their bond order. In the quest for the limits of FM-AFM resolution, we turn into the Moire structures formed upon the epitaxial growth of graphene on metals.

Understanding the local modulations in the structure, charge density and electronic properties of graphene induced by the interaction with the metal is currently an active field of research due to its possible technological applications. The low interacting cases, like graphene on Pt(111), are of utmost interest as the most characteristic property of graphene, the linear dispersion around the Dirac point, is preserved. This low graphene-metal interaction results also in moiré patterns with small topographic corrugation. Whereas STM easily resolves the moirés through its dependence of the local density of states (LDOS) [1], this system represents a real challenge for FM-AFM and an ideal test bed for examining the sensitivity of the technique to almost pure local electronic variations.

In this talk, we'll focus on the theoretical analysis of recent FM-AFM images on the graphene/Pt(111) system taken with a home-made AFM operating at 5K and UHV [2]. These studies have resolved different moiré patterns and shown an inversion of atomic contrast from an hexagonal to a triangular lattice as a function of the distance. Our first-principle calculations explore in detail the interplay in the image contrast formation between the chemical bonding and the van der Waals (vdW) interaction. The latter is included with different approaches ranging from semi-empirical methods to exchange-correlation functionals that include the non-local correlations responsible for the vdW forces. We show that the inversion of atomic contrast can be rationalized in terms of the electronic density dependence of the Pauli interaction [3]. Regarding the moirés, a subtle contrast may be observed for very reactive tips in the attractive regime. However, we associate the common AFM observation of the moiré to sub-surface resolution. The non-topographic corrugation of the moiré is obtained in the repulsive regime, where the tip indents the graphene sheet deep enough so the displaced carbon atoms act as a tip that allows sensing the Pt surface with atomic resolution. This idea may be generalized to other 2D materials opening the door to simultaneous monolayer/substrate AFM characterization.

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Direct nanopatterning of atomically-thin TMDCs conducting layers via local oxidation nanolithography

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Since the discovery of graphene, a great deal of research efforts have been made to integrate atomically thin layers in the fabrication of ultrathin, transparent and flexible electronic devices [1] The fundamental advance in this area has been feasible thanks to the micro-mechanical exfoliation methods.[2] In this context, 2D crystals such as transition metal dichalcogenides (TMDCs) have shown excellent exfoliation mechanism and offer a wide variety of properties that could match or even outweigh the properties of graphene. In this work, it will be reported the controlled chemical modification of ultrathin TaS_2 layers by means of AFM Local Oxidation Nanolithography (AFM-LON) to produce two-component surfaces formed by metallic TaS_2 layers and insulating oxide motifs.

TaS_2 crystals were exfoliated by a novel technique based on the controlled application of shear forces between the 2D crystal and the substrate. The TaS_2 atomically-thin layers, which are metallic at room temperature and superconducting at low temperatures, were conveniently nanopatterned with an unprecedented precision and reproducibility only achieved thanks to the development of a new LON modality, named by us as *static-tip* LON. It was finally extended to other conducting TMDCs proving the versatility of this technique.

The possibility of the controlled nanopatterning of atomically-thin TMDCs layers opens the door for the nanofabrication of devices and furthermore, for the fundamental study at low temperature of the physical consequences derived from the confinement at the nanoscale of these layered superconductors.

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Figure

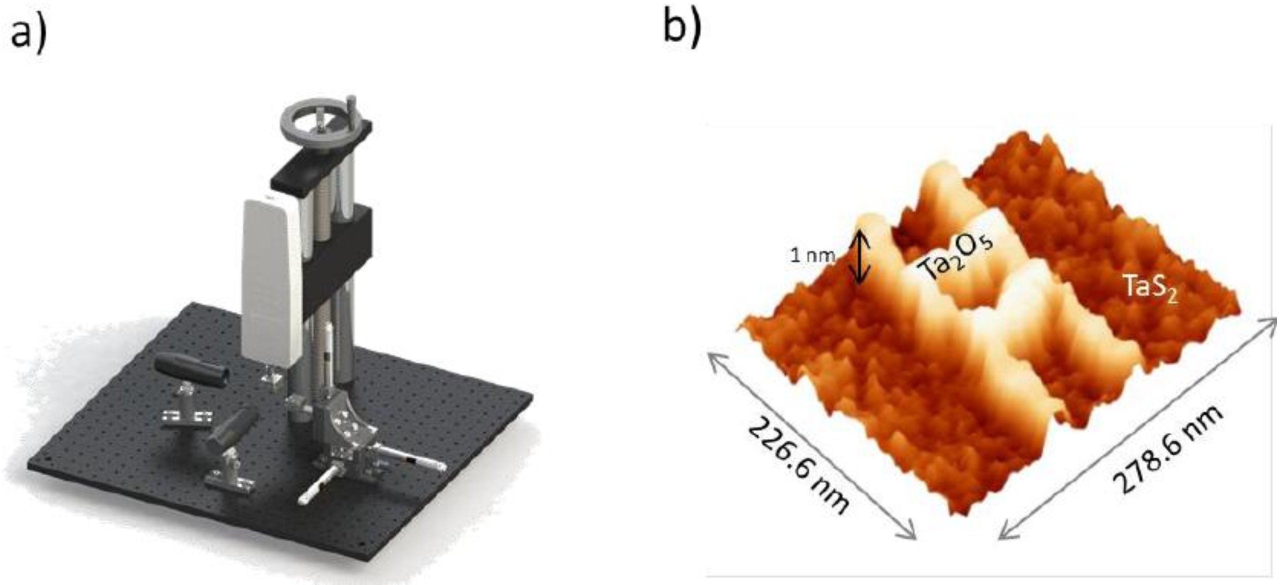

Figure1: a) 3D rendered view of the set-up used for the 'press and shear' exfoliation of TMDC single crystals. b) Fabrication of the active part of a single electron transistor (SET) out of a TaS_2/Ta_2O_5 structure generated by Static-tip AFM-LON. The island on the center of the pattern is isolated by lines of 1 nm high of Ta_2O_5 that could act as tunnel junctions.

Electrostatic Exfoliation of Graphene on Graphite

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The purpose of this study is to investigate the exfoliation of graphene from a graphite surface using a Scanning Tunneling Microscope (STM). The major objective of this research is to demostrate that this can be done without touching the graphite surface, just with the electrostatic interaction between the metalic tip of the STM and the graphite sample.

A study of the different parameters that could influence the exfoliation mechanism was performed, such as the bias voltage applied between tip and sample, the effect of the graphite terraces edge structure, tip geometry and the enviromental atmosphere. Finally, Density Funciontal Theory (DFT) calculations were carried out in order to provide an understanding of the interactions between layers in graphite.

On the basis of the results of this research, it can be concluded that the mechanism of the electrostatic exfoliation of graphene is a result of different effects: on the one hand, the electrostatic force decouples the two last graphite layers and breaks atomic bonds, and as a second stage the electrostatic interaction between STM tip and graphite sample during the scanning of the surface generates graphene flakes.

In the future it would be interesting to further study the effect of the polarity of the applied voltage between tip and sample and to provide an exfoliation mechanism that does not depend on the STM tip scanning action

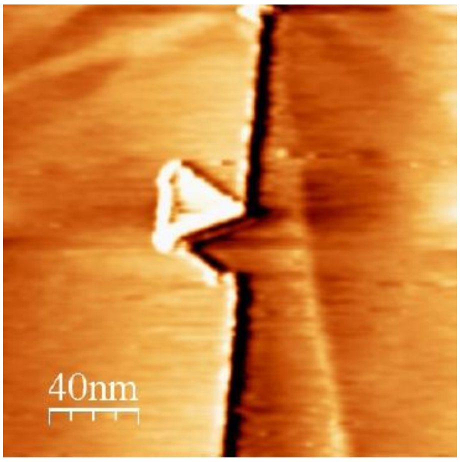

Figure1: Monoatomic graphite terrace after voltage ramp showing graphene flake.

Physical concepts behind electrostatic and magnetic force microscopy

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In this tutorial I will briefly review basic concepts of electrostatic and magnetic interactions relevant to understand long range forces in scanning probe microscopy (SPM). We will discuss the field distributions around sharp conducting and magnetic tips. Based on very simple models, we will show how to estimate electrostatic and magnetic force and force gradients in typical SPM set-ups.

Microscopy and Spectroscopy under ambient gas and liquid environments

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This tutorial will cover the principles and the operation of Surface sensitive techniques that operate under gases near atmospheric pressure and under liquids. They expand the last decade's advances in our fundamental understanding of materials surfaces brought about by the application of such tools as Auger and Photoelectron Spectroscopies, Scanning Probe Microscopies and Scanning and Transmission Electron Microscopies, which operate only in high vacuum. They have served well the electronic, chemical, catalysis and environmental industries. Examples include the discovery of the mechanism and the nature of active sites in ammonia synthesis, the discovery of the unique catalytic properties of Au nanoparticles for CO oxidation, the discovery of the nature of the active sites for hydro-desulfurization in MoS_2 nanoparticles, and many more.

The situation regarding solid-gas and solid-liquid interfaces is far from this level of understanding and success. The reason for this is that the powerful surface sensitive techniques used in the past do not work well under gas and liquids at or near atmospheric pressures. This tutorial course will review the development and operation of new techniques and methods that remedy this. We will particularly review the operation of photoelectron spectroscopies under gas pressures in the tens of Torr range. Most common and widely used is X-ray Photoelectron Spectroscopy or XPS. We then will review X-ray absorption spectroscopy (XAS, also known as NEXAFS), and x-ray emission spectroscopy (XES). These two techniques involve the absorption and emission of photons and thus are not intrinsically surface sensitive as the mean free path of photons is much larger than that of electrons. However, XAS can be made surface sensitive by collecting secondary emitted electrons instead of fluorescence photons. We will also see how in this way the electrode-electrolyte interface can be studied in situ. We will discuss new properties and new phenomena that have been discovered by their application in the area of environmental science, electro-chemistry and catalysis.

Charge transport in PbS quantum dot arrays for photovoltaic applications

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Charge hopping and percolation in disordered semiconductors has been widely studied, but the microscopic nature of the percolation process is not understood and has never been determined. Here we directly image the charge percolation pathways in 2-D PbS quantum dot (QD) arrays using Kelvin probe force microscopy. We show that the electrons percolate via in-gap states (IGS) instead of conduction band states, while holes percolate via valence band states. This exotic transport behavior is explained by the electronic structure and energy level alignment of the individual QDs, which was measured by scanning tunneling spectroscopy. Density functional theory and spectroscopic analysis show that the IGS are induced by oxygen molecules adsorbed on the QDs' surface. These states are partially hybridized with the valence band states, enabling inter-IGS coupling and electron transport via IGS. Surface chemical treatments open the way of defect engineering, achieving tunable electronic structure and transport properties by controlling the IGS.

Polarization-Resolved Near-Field Mapping of Nanoscale (λ_0/220) IR Transmission Line Modes

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Metal antennas and transmission lines (TL) are common devices for receiving and transporting signals in the radiofrequency regime. It has been demonstrated that by reducing the size down to the micrometer range, these devices can be operated at infrared frequencies (~30 THz) [1-2].

Here we demonstrate that functional infrared TLs with gap widths down to 25 nm can be fabricated by Focused Ion Beam (FIB) milling of gold films on CaF_2 substrates. Interferometric and polarizationresolved near-field microscopy [3] is applied to map in real space the propagation of the TL modes. For the first time, we measured the strongly confined fields of a propagating TL mode by mapping the spolarized scattered field (Fig. 1) [4]. Imaging TLs with 25 nm gap width we experimentally demonstrate an infrared mode with diameter of Dm = 42 nm (λ_0/220), which intriguingly, shows a propagation length of about L_m = 8 µm. Interestingly, this is more than two orders of magnitude larger than the mode diameter, L_m/D_m = 190. It is worth to be noted that the mode wavelength λ_m for 25 nm gap is still in the micrometer range, showing that nanoscale field confinement in infrared TLs does not require wavelength compression to the nanometer scale. The mechanism of field confinement in the TLs is rather of geometrical nature. The two metal wires are the mode carriers, and the field confinement is achieved by capacitive coupling between the TL wires. Infrared TLs are thus interesting candidates for the development of ultra-compact infrared bio-chemical waveguide sensors or circuits.

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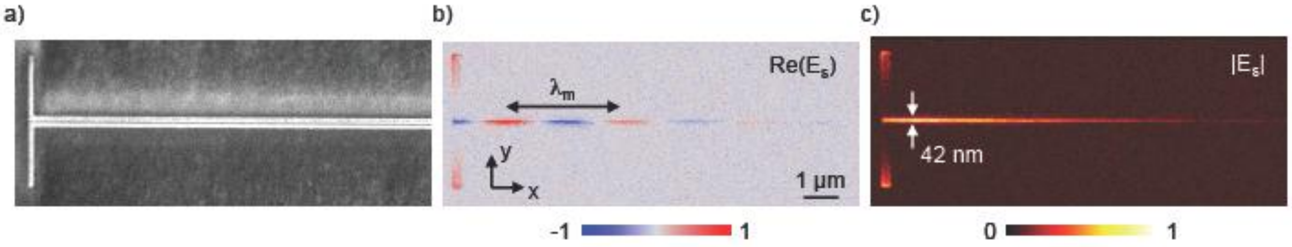

Figure1: Near-field imaging of a TL with a 25 nm gap width by detecting s-polarized scattered field, E_S. a) SEM image of the TL. b) Near-field image showing the real part of the s-polarized scattered field, R_e(Es). c) Near-field image showing the real part of the s-polarized scattered field, $|E_s|$.

Quantum states and engineering dangling bond structures in silicon

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The scanning tunnelling microscope (STM) first demonstrated its potential for application in the creation of quantum structures some twenty years ago with the construction of the first quantum corral which illustrated bound electron states extending across multiple lattice sites on the surface [1]. Here, we form extended quantum bound states on the surface by coupling individual atomic orbitals. The resultant structures can be considered as analogous to molecular orbitals. By selective desorption of individual hydrogen atoms from a hydrogen-terminated Si(001) surface using the STM tip, we can create dangling bond (DB) states in precisely chosen locations on the surface lattice. We make small chains of DBs over adjacent lattice sites and then vary the STM tip bias in imaging, seeing contributions to the images from both the ground and excited states of the resultant "molecular" quantum bound states as illustrated in Figure 1 [2]. We further seek to generalise our findings by examining dangling bond states on a wider range of silicon surface facets. The excited states we image provide a new perspective on semiconductor defects and are relevant to the wider challenge of quantum state engineering through the atomically precise manipulation of chemically passivated semiconductor surfaces.

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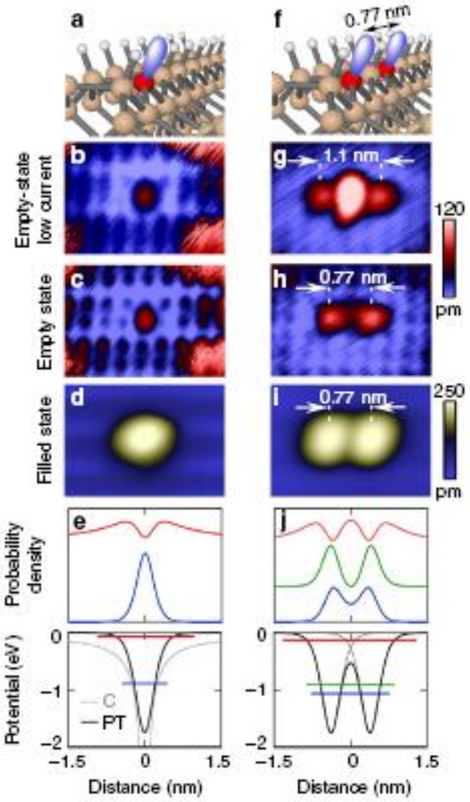

Figure1: Illustrating a single dangling bond (a) and pair (f) imaged in both the filled (d, i) and empty states (b-c, g-h). An unexpected "molecular" excited state is seen for the pair at low currents in the empty states (g) and is explained in the context of extended quantum bound states (e, j).

Ultrathin MgO/Ag(100) films: how the final morphology depends on growing conditions and interfacial oxygen.

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The ability to engineer nearly perfect ultrathin oxide layers, up to the limit of monolayer thickness, is a key issue for nano-technological applications in fields as catalysis, corrosion protection, micro- and nano-electronics, sensoristics, spintronics, drug delivery, etc. [1]. We faced the difficult and important case of ultrathin MgO films on Ag(100), for which no extended and well-ordered layers could so far be produced in the monolayer limit. The stoichiometric and morphological characteristics of MgO monolayers were shown to depend critically on growth parameters as temperature (T_g) [2], O_2 partial pressure and Mg evaporation rate [3] during reactive deposition. However, contrasting data are present in literature with respect both to the optimal T_g [2,4] and to the island morphology and orientation [5,6]. Combining STM, XPS and HR-EELS experiments we demonstrate that the structure of sub-monolayer MgO films grown on Ag(100) by reactive deposition is strongly affected not only by the growth conditions but also by after-growth treatments. The latter ones allow to quench the thermodynamically most stable configuration at T_g or to let the system evolve towards the low temperature equilibrium state [7]. Moreover, at T_g=773 K the films show an excess of oxygen, suggesting that oxygen atoms accumulate at the MgO/Ag interface. The presence of these interface oxygen atoms reduces the stress of the oxide film favouring the formation of extended terraces. Preliminary DFT calculations confirm this experimental evidence [8]. The result is the possibility to tune the morphology of the films from small islands with corrugated borders, to perfectly square islands of larger size to MgO terraces tens of nm wide (see Fig. 1). Since the film structure influences both chemical and electronic properties [1] of the layers, a full control of all experimental parameters opens important perspectives for applications in catalysis and for the use of ultrathin oxide films as support for the further deposition of organic and inorganic nano-objects.

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Figure

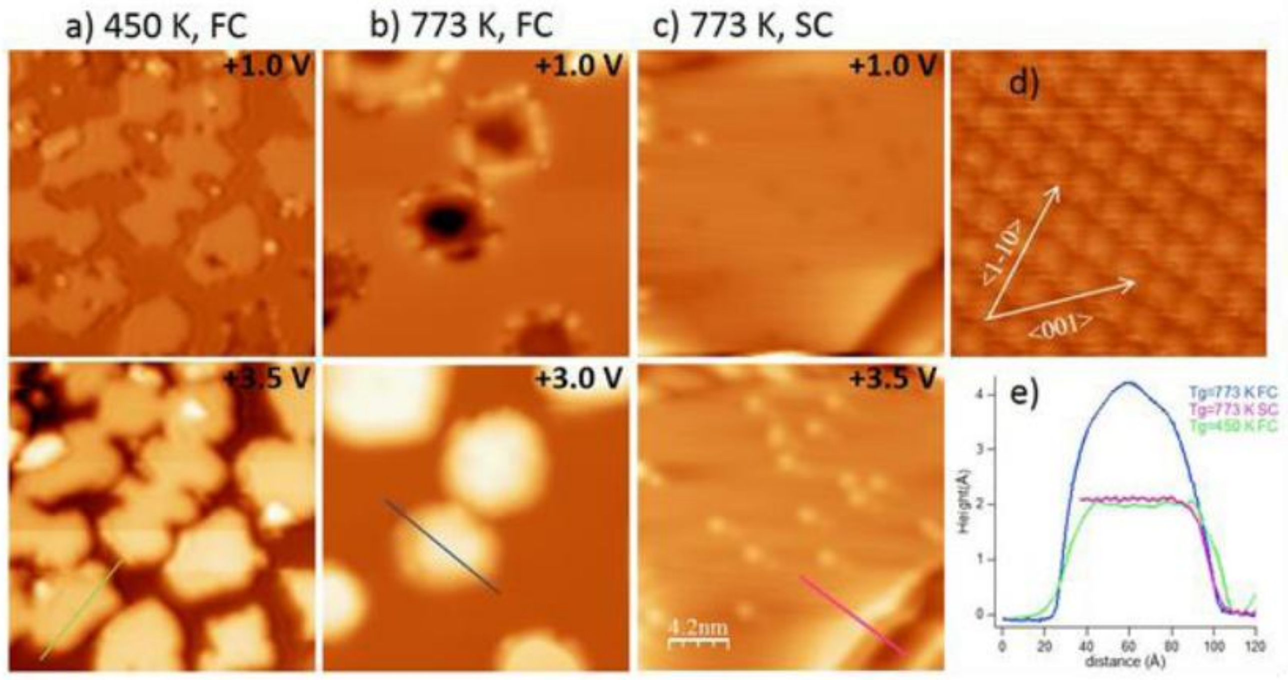

Figure1: Columns (a-c): STM Images of MgO films of 0.7 ML nominal thickness grown under different conditions. (a) T_g=450 K and fast cooling below room temperature (FC); (b) T_g=773 K, FC; (c) Tg=773 K, and slow cooling (~5 K/min - SC). For all panels, image size 21x21 nm^2; I=0.2 nA. Panel (d): Atomically resolved image of clean Ag(100), used for calibration. High symmetry directions are marked by arrows. Image size: (2.4x2.4) nm^2, V=0.1, I=0.2 nA. Panel (e): height profiles of the different MgO structures cut along the lines marked in the bottom panels (a-c) (<001> direction, topographic conditions). It is evident from these images that a careful control of both growth and after growth parameters allows to tune the formation of irregular monolayer islands, of squared bilayers or of extended monolayer terraces.

Non-collinear spin texture of fct-Mn on Co/Cu(001) at room temperature

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Mn, as an antiferromagnetic 3d metal, has drawn much attention in ultra-thin magnetic film research in the last decades, not only because of its large atomic magnetic moment due to the half-filled 3d orbital, but also the diversity of phases when epitaxially grown on different substrates [1-5]. In this contribution, face-centered-tetragonal (fct) Mn on Co/Cu(001) was studied by scanning tunneling microscopy (STM) and spectroscopy in connection with the lock-in technique at room temperature. A spin-polarized bulk iron ring probe with in-plane magnetization [6] was employed for scanning, fitting to the expected in-plane orientation of the magnetization of the antiferromagnetic Mn in this system [7].

5.4 atomic monolayers (ML) Mn were evaporated on 4.0 ML Co/Cu(001) by molecular beam epitaxy in vacuo. A local differential conductance map was recorded simultaneously with the topography during the constant-current-mode STM scan on 6-ML terraces of the Mn film. Both are shown in Fig. 1(a). In the topography image, a corrugated surface with two orthogonal domains in the left and the right part of the image is observed. Both show the same periodicity, as demonstrated for one direction by the line scans 1 and 2 in the bottom of Fig. 1(c). The corrugation amounts to up to about 0.3 Å, which could come from an outward buckling of Mn atoms on the mismatched substrate [3,4]. A (12×2) unit cell can be identified on the surface structure, as highlighted by the yellow rectangles. The low-energy electron diffraction (LEED) pattern showed only sharp (1×1) spots with a somewhat enhanced diffuse background compared to Cu(001) and Co/Cu(001) (Fig. 1(b)), which could mean that the domains with this superstructure are smaller than the coherence length of the LEED system. While the topography of the two domains is the same, just rotated by 90°, the dI/dV contrast shows notable differences. The left domain exhibits a different periodicity, as evidenced by the line scans in Fig. 1(c). Line scan 1 (left area) in the dI/dV map has half the periodicity of line scan 2 (right area), and also half of the topographic periodicity of line scan 1. We interpret this as the effect of spin contrast, where the orientation of the tip magnetization breaks the symmetry present in the topography image. The orientation of the iron ring probe is illustrated by the black arrow in the dI/dV map in Fig. 1(a). The magnetic moment of the apex atom should be along the tangential direction of the ring. Since this magnetic moment has a larger component along the [110] direction, the contribution from spin contrast should be dominated by the spin component of Mn atoms along this direction. We thus conclude that the Mn/Co/Cu(001) system presents a non-collinear surface spin structure with a (12×2) periodicity at room temperature.

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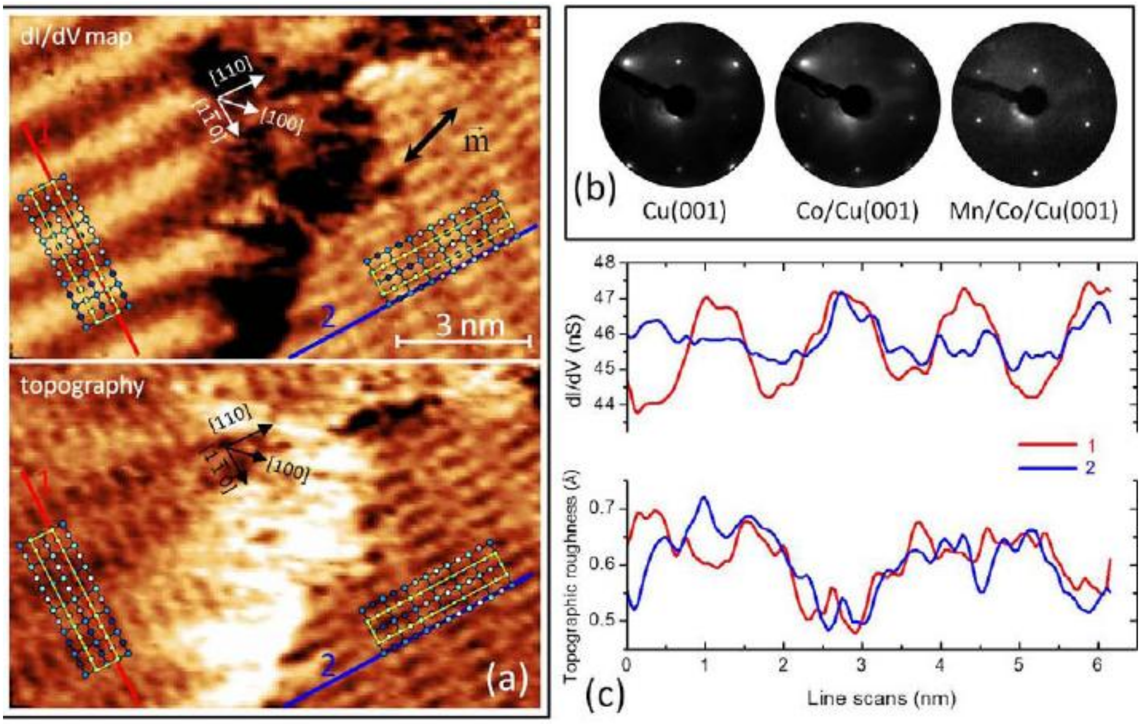

Figure1: (a) dI/dV map (top) and topography (bottom) of Mn surface, gap voltage 0.2 V, tunneling current 10.5 nA (b) LEED images at 117.8 eV for Cu(001) (left), 4 ML Co/Cu(001) (middle) and 5.4 ML Mn/4.0 ML Co/Cu(001) (right) (c) Line scans of (a) along two orthogonal domains in both the dI/dV map (top) and topography (bottom).

Propagating plasmons in carbon nanotubes for nanophotonic circuits

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Carbon nanotubes (CNTs) offer interesting nanophotonic and optoelectronic applications. Supporting plasmons with extremely short wavelengths, they provide a strong light compression critical for the development of future miniaturized optical devices. Here, we propose carbon nanotubes as efficient coupling and launching elements for nanophotonic circuits. By full-wave analysis we demonstrate the manipulation of propagating plasmons along CNTs and their coupling to plasmons in graphene resonators. We also show an efficient way of launching and phase control of propagating graphene plasmons with the help of a CNT antenna. Combining CNTs with graphene could become a promising route towards mid infrared plasmon circuitry.

Previously, plasmons in graphene have been visualized by scanning near-field optical microscopy (scattering-type SNOM) [1-3]. SNOM presents an atomic force microscope in which a metallic tip is illuminated with a focused infrared laser beam. The backscattered radiation is recorded simultaneously with the topography, yielding nanoscale resolved infrared near-field images. The propagating plasmons in CNTs have still not been observed by any experimental technique. We foresee the SNOM to be an efficient tool for proving the existence of plasmons in CNTs and testing future design concepts for CNT nanoscale plasmonics.

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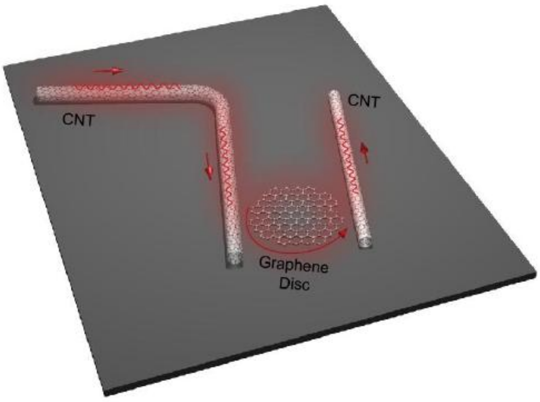

Thin film nano-morphology analysed by nfrared near-field microscopy and spectroscopy

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Using a scattering-type scanning near-field optical microscope (s-SNOM) our group analyses nano-structures by near-field imaging and nano-FTIR.1 We are able to resolve shifts in vibrational modes of small molecular crystals according to different packing structures. The spatial resolution is only limited by the tip apex of around 20 nm, which is far better than the common infrared far-field analysis methods. The spectral resolution is 2 cm^{-1}. We find that state-of-the-art pentacene films grown on SiO_2 at elevated temperature are structurally inhomogeneous consisting of two interpenetrating phases. We observe bulk-phase nucleation (of distinct ellipsoidal shape) on a submicron scale within the dominant pentacene thin-film phase that even proceeds with storage time.

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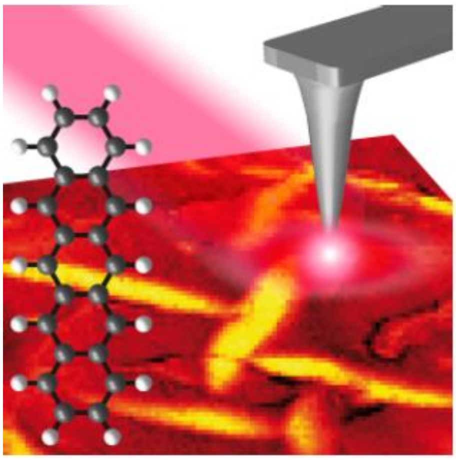

Figure1: Ongoing work with the s-SNOM concentrates on improving the signal to noise ratio of the nano-FTIR technique, and the ability to perform hyperspectral imaging

Atomically Resolved Graphitic Surfaces in Air by Atomic Force Microscopy

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Imaging at the atomic scale using atomic force microscopy in biocompatible environments is an ongoing challenge. We demonstrate atomic resolution of graphite and hydrogen-intercalated graphene on SiC in air (Fig. 1) [1]. The main challenges arise from the overall surface cleanliness and the water layers which form on almost all surfaces. To further investigate the influence of the water layers, we compare data taken with a hydrophilic bulk-silicon tip to a hydrophobic bulk-sapphire tip. While atomic resolution can be achieved with both tip materials at moderate interaction forces, there are strong differences in force versus distance spectra which relate to the water layers on the tips and samples. Imaging at very low tip-sample interaction forces results in the observation of large terraces of a naturally occurring stripe structure on the hydrogen-intercalated graphene. This structure has been previously reported on graphitic surfaces that are not covered with disordered adsorbatesin ambient conditions (i.e., on graphite and bilayer graphene on SiC, but not on monolayer graphene on SiC). Both these observations indicate that hydrogen- intercalated graphene is close to an ideal graphene sample in ambient environments [1].

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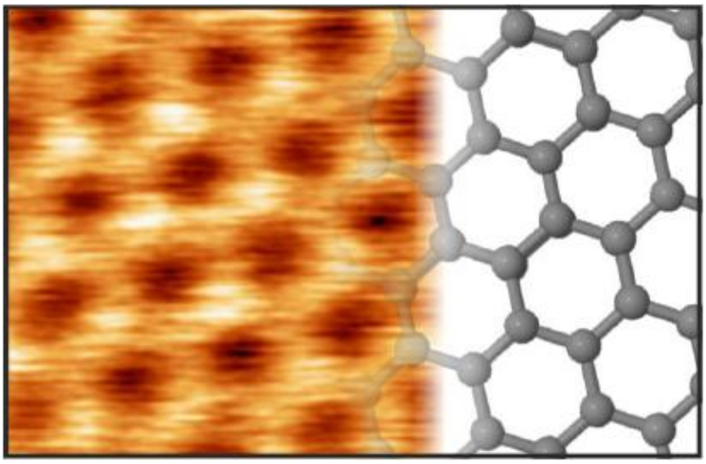

Figure 1: Atomic resolution on HOPG in air compared with structural model. Scan and sensor parameters: qPlus sensor with silicon tip, f_0 = 29858 Hz, k=1280N/m, Q_{air} =2432 , Δf = 200 Hz, A= 380 pm.

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