Spin dynamics in single molecule magnets

Quantum tunneling of magnetization and quantum phase interference have been intensively studied in single molecule magnets (SMMs), high spin molecules with an easy axis (Ising) type magnetic anisotropy. SMMs have also been proposed as candidate qubits—and very recently several groups have reported the observation of coherent spin dynamics in pulsed EPR studies. In the first part of this talk I will review experiments and theory of SMMs, including the spin-Hamiltonians that capture the physics of their spin-dynamics. I will then discuss recent experiments in my group on spin-relaxation between low-lying spin-states in Ni4, an S=4 SMM [1, 2].


In an effort to produce new examples of single-molecule magnets, we are exploring directed assembly routes to high-nuclearity metal-cyanide clusters. The use of multidentate capping ligands has led to the synthesis of a range of polygonal and polyhedral species featuring as many as 27 metal centers. Substitution of a variety of transition metal ions into these geometries is shown to permit adjustment of the ground state spin and magnetic anisotropy associated with the molecules. In particular, the use of second- and third-row transition metal ions with a large spin-orbit coupling is found enhance magnetic relaxation barriers. Attempts to extend this approach to symmetric bridging ligands facilitating strong electronic coupling will also be described.


A molecular magnet is composed of a collection of six-fold coordinated spin-polarized transition-metal ions that are held together by ligands. When exchange interactions between neighboring metal centers are strong enough, the molecule can be viewed as being in a nearly classical configuration consisting of a single spin-ordered ground-state with a high degeneracy. Inclusion of the spin-orbit interaction breaks this degeneracy at second order. From a technological standpoint, an interesting case occurs when the molecule has uniaxial symmetry as this leads to a case where the energies of the lowest spin manifold can be characteristic of a classical bar magnet. However it is well established that these nanoscale molecules still behave as a quantum magnet and that the $2S+1$ magnetic states are split into a collection of doublets and one singlet. Resonant tunneling of magnetization between these states has been observed and is now well established.

At the nanoscale, interactions between a molecular magnet and its surroundings affect the collective magnetic behavior of the molecule. Possible effects that have been experimentally observed and/or computationally investigated include: (1) dramatic changes in spin ordering or magnetic anisotropy barriers due to the addition of one or two electrons, (2) enhanced tunneling rates or the appearance of tunnel-splitting oscillations (Berry's Phase) due to weak interactions between the molecule and space-filling spectators (such as H$_2$O), (3) changes in the magnetic anisotropy Hamiltonian due to effects such as pressure or physisorption of the molecular magnet on a surface/electrode. Due to all of these effects, it is reasonable to expect that a many-body approach may be necessary to reliably explain the current-voltage characteristics associated with electronic transport across molecular magnets. Recent theoretical reviews can be found in Refs, [1-2]
With the Mn12-Acetate molecule as a concrete example, I will discuss recent work aimed at developing a compact many-electron and many-spin representation of molecular magnets as a function of charge/excitation and applied electric and magnetic fields. A recent application of this theoretical and computational method will be presented as well (L. Michalak, C. Canali, M.R. Pederson, M. Paulson, and V. Benza, Submitted to PRL See also: http://arxiv.org/PS_cache/arxiv/pdf/0812/0812.1058v1.pdf)

Earlier pstdy of this work were supported by ONR, NSF and NRL core funding while MRP was at NRL. MRP thanks NRL and NRL CCMS for continuing to providing the requisite resources for performing the recent parts of this work. Computational support was provided by the DOD-HPCMO.


We determine the effect of an in-plane current flow on the critical properties of a 2d itinerant electron system near a ferromagnetic-paramagnetic quantum critical point. We study a model in which a nonequilibrium steady state is established as a result of exchange of particles and energy with an underlying substrate. Current flow $j$ has two important effects. One is to give rise to decoherence or an effective temperature which causes a crossover from quantum to classical behavior. The second effect of current flow is to give rise to symmetry breaking terms of the form $\vec{j} \cdot \vec{\nabla}$ in the effective action which qualitatively has the effect of producing a drift of the order-parameter. The effect of the symmetry breaking on the fluctuational and critical properties is found to be small near the quantum critical point, with the dominant effect being that of current induced decoherence. In the classical ordered regime, the effect of the symmetry breaking terms are found to be far more important. Here they may be identified as current-induced spin torque terms in the classical Landau-Lifshitz-Gilbert equations which can make the uniformly ordered state dynamically unstable.

Molecular spins on the road of quantum computation

Molecular magnets have proven key quantum effects - like quantum tunneling of the magnetization and Berry phase interference – originating from their well mastered structure. Nowadays a growing interest is dedicated to the physics and chemistry of such systems. The appealing fundamental quantum properties of the magnetic molecules – and in general of isolated quantum spins – have brought them into the race of becoming viable candidates for future quantum bits. In this lecture I will review some of the low-temperature measuring techniques and studies realized on molecular magnets and how they compare to classical nanomagnets. Prospects on how to use NMR-like pulse techniques to control the spin dynamics will be discussed, in analogy with recent experiments realized on superconducting quantum bits. Recent experiments at NHMFL achieving coherent spin manipulation will be shown. We have used large spin Mn2+ ions (S=5/2) to realize a six level system that can be operated by means of single as well as multi-photon coherent Rabi oscillations. This spin system has a very small anisotropy whose effect can be tuned in-situ to turn the system into a multi-level harmonic system. This offer new ways of manipulating, reading and resetting a spin qubit. Decoherence effects are strongly reduced by the quasi-isotropic electron interaction with the crystal field and with the 55Mn nuclear spins.

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<th>Irinel Chiorescu, Florida State University</th>
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<td>Molecular spins on the road of quantum computation</td>
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<td>When</td>
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<td>Description</td>
<td>Molecular magnets have proven key quantum effects - like quantum tunneling of the magnetization and Berry phase interference – originating from their well mastered structure. Nowadays a growing interest is dedicated to the physics and chemistry of such systems. The appealing fundamental quantum properties of the magnetic molecules – and in general of isolated quantum spins – have brought them into the race of becoming viable candidates for future quantum bits. In this lecture I will review some of the low-temperature measuring techniques and studies realized on molecular magnets and how they compare to classical nanomagnets. Prospects on how to use NMR-like pulse techniques to control the spin dynamics will be discussed, in analogy with recent experiments realized on superconducting quantum bits. Recent experiments at NHMFL achieving coherent spin manipulation will be shown. We have used large spin Mn2+ ions (S=5/2) to realize a six level system that can be operated by means of single as well as multi-photon coherent Rabi oscillations. This spin system has a very small anisotropy whose effect can be tuned in-situ to turn the system into a multi-level harmonic system. This offer new ways of manipulating, reading and resetting a spin qubit. Decoherence effects are strongly reduced by the quasi-isotropic electron interaction with the crystal field and with the 55Mn nuclear spins.</td>
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The concept of classical magnetism relies on continuous angular orientation of the magnetization. If magnetic structures shrink to a level at which they contain only a few coupled atoms, they become quantum spin systems that have only discrete possibilities for their orientation of magnetization. These nanomagnets allow fundamental insight into basic principles of quantum mechanics. At the same time they evoke high interest as possible candidates for miniature data storage.

We use a scanning tunneling microscope (STM) operating at low temperature and in high magnetic fields to probe electron spin systems of transition metal atoms such as Fe, Co or Mn atoms on well characterized adsorption sites on a one monolayer thin Cu2N layer on Cu (100). Atom manipulation is used to construct magnetic nanostructures one atom at a time. The adsorbed atoms are incorporated into the extended molecular structure of the Cu and N atoms through polar covalent bonds and couple strongly to each other forming a coherent spin system. Inelastic spin excitation spectroscopy measures the energies connected to excitations of this spin. The observed spin transitions are well described within the framework of a Heisenberg-type Spin Hamiltonian accounting for magneto-crystalline anisotropy and magnetic exchange interaction between coupled atoms. By attaching one transition metal atom to the direct apex of the STM’s tip we gain an additional possibility to interact with the surface-bound spin system. The distance between tip spin and magnetic structure on the surface becomes a continuously adjustable parameter. This results in a tunable spin-spin coupling, as opposed to the discreet structural configurations in the surface-bound atoms. The spin excitations are strongly dependent on the spin of the tunneling electrons which is detectable by the spin-filtering properties of the tip-bound atom. The combination of spin resolving measurements with the ability to vary magnetic coupling at the atomic level enables...
further exploration of quantum magnetism.


Stuart Parkin, IBM

Spin torque driven dynamics of magnetic domain walls in nanowires

When
Mon Jan 12 7:05pm – Mon Jan 12 7:35pm

Created By
Aspen Winter Conference on Unifying Themes in Condensed Matter

Description
New paradigms for domain wall (DW) based logic and memory devices are made possible by the direct manipulation of DWs using spin polarized electrical current via the phenomenon of spin transfer torque. We have recently demonstrated the controlled motion of a series of up to 6 vortex domain walls, back and forth, along nanowires formed from permalloy (Ni81Fe19) using nanosecond long current pulses. This is not possible using magnetic field in which adjacent DWs are driven in opposite directions. The manipulation of DWs via spin torque is the fundamental principle governing the Racetrack Memory, a novel storage-class memory device [1,2]. Using both quasi-static and time resolved measurements of the resistance of permalloy nanowires we have explored the detailed interaction of spin polarized current with DWs with different structures and their transformation between these structures induced by this interaction. In particular, the role of pinning sites, whether intrinsic or extrinsic, can fundamentally change the nature of this interaction allowing for the resonant excitation of DWs [3,4].


[3] L. Thomas et al., 443, 197-200 (2006);

### What
Seamus Davis, Cornell University and Brookhaven National Laboratory  
*Quasiparticle Interference Imaging of Cuprate Electronic Structure in the Superconducting and Pseudogap States*

### When
Tue Jan 13 8:15am – Tue Jan 13 9:15am

### Created By
Aspen Winter Conference on Unifying Themes in Condensed Matter

### Description
We will review the advances in visualization and understanding of the electronic structure of cuprate superconductivity using Fourier transform scanning tunneling spectroscopy (FTSTS) to image quasiparticle interference effects. A comprehensive and consistent empirical picture of the $T \rightarrow 0$ electronic structure emerges. It reveals a fundamentally bipartite electronic structure with heterogeneous quasi-localized high-energy states near $k \sim (0, \pm \pi/a_0)$ which are dominated by dopant-induced electronic disorder, and spatially homogeneous low energy k-space states which are demonstrably the excitations of Cooper pairs. We explore the evolution of all these phenomena as the carrier density of hole-doped cuprates is reduced from the robust high temperature d-wave superconductor towards the non-superconducting Mott insulator phase.

We also use high precision FTSTS to visualize the momentum-space excitations in the mysterious ‘pseudogap’ phase occurring at $T > T_c$ in the underdoped cuprates. We report the first studies of the continuous temperature dependence of the complete ‘octet’ of quasiparticle interference signals from $T < 0.1T_c$ to $T > 1.25T_c$. A comprehensive and detailed understanding of all states with $E < 40$meV and for a full reciprocal unit cell is achieved. Using these data, we identify the low temperature state of the cuprate ‘pseudogap’ regime which dominates transport and quantum oscillation phenomena. Finally we explore the distinct identity of the higher energy excitations which dominate the high temperature thermodynamic characteristics of the ‘pseudogap’ state in these materials.

We discuss several important conclusions about the mechanism of superconductivity and the limitations on the critical temperature.
Gregory Boebinger, National High Magnetic Field Laboratory

Quantum Oscillations in the Specific Heat of Ultraclean YBCO in 45T magnetic fields.

What
Gregory Boebinger, National High Magnetic Field Laboratory

Description
We report the observation of quantum oscillations in specific heat measurements of ultra-clean YBCO_6.5 in a magnetic field as large as 45T, allowing a direct comparison of the fully superconducting state with the normal (or mixed) state at 45T and low temperatures (T/Tc < 0.05). This thermodynamic measurement of the electronic density of states determines the total number of carrier pockets in the two-dimensional Fermi surface of the cuprates. Quantum oscillations provide a bulk measurement of the quasiparticle density of states in the d-wave superconducting cuprates. Jokes will be told.
Suchitra Sebastian, Cambridge UK

Quantum Oscillations in high Tc's: the nature of Fermi surface reconstruction and relation to the Mott insulating regime

I will present results of quantum oscillation measurements we have performed in the underdoped cuprate YBCO6+x and the parent pnictide SrFe2As2, providing evidence for a small Fermi surface. Potential parallels are drawn between the nature of underlying order that reconstructs the Fermi surface in these two families of materials. Differences, however are highlighted in the nature of the parent material in these two cases - a series of doping-dependent measurements in YBCO6+x will be presented that trace the Fermi surface evolution as the insulating regime is approached.
Following the discovery of long-range antiferromagnetic order in the parent compounds of high-transition-temperature (high-Tc) copper oxides, there have been efforts to understand the role of magnetism in the superconductivity that occurs when mobile ‘electrons’ or ‘holes’ are doped into the antiferromagnetic parent compounds. Superconductivity in the newly discovered rare-earth iron-based oxide systems RFeAsO (R, rare-earth metal) and Sr(Ba)Fe2As2 also arise from either electron or hole doping of their non-superconducting parent compounds. Elastic neutron scattering was used to demonstrate that static collinear antiferromagnetic order is the ground state of undoped parent compounds of all FeAs-based high-Tc superconductors [1-5]. The magnetic structure and its doping dependence were determined for several systems, showing that when superconductivity sets in, static antiferromagnetic order is eliminated. We use inelastic neutron scattering to study spin excitations in superconducting BaFe1.9Ni 0.1As2 and spin waves in (Ca,Sr)Fe2As2 [6,7]. Our results shed new light to the interplay between magnetism and superconductivity in these FeAs-based superconductors.

Hai-Hu Wen, Chinese Academy of Sciences

Unconventional superconductivity and mixed state properties in FeAs-compounds

Using flux method we have successfully grown the NdFeAsO1-xFx and Ba1-xKxFe2As2 single crystals with high quality. It is found that the anomalous electron scattering in the normal state cannot be simply attributed to the multiband effect. The influence given by the magnetic correlation/scattering may play an important role. Specific heat, lower critical field and point contact tunneling measurements have been carried out and all indicated the unconventional superconductivity and multigap features. While the paring symmetry of the superconducting gap may be a non-trivial issue. In the electron-doped 1111 phase, the superfluid density is rather low and possesses probably a nodal feature. The specific heat anomaly C/T|Tc is about 5-8 mJ/mol-Fe-K2 which is close to the predicted value of the band structure calculations. While in the 122 phase, C/T|Tc is as large as 50 mJ/mol-Fe-K2 which is 5-10 times higher than that in the 1111 phase. Also the superfluid density in 122 phase is about 5-10 times higher than that in the 1111 phase. An s-wave component was found in the 122 phase by low temperature specific heat measurements, but the low temperature penetration depth measurements reveal a clear nodal feature even in the 122 phase. All these results together suggest that the gap on the hole pocket may be s-wave like, while that near the M point may have nodes. Superconductivity based on spin fluctuation mediated mechanism will be discussed.

By measuring magnetization and resistivity, we have determined the anisotropy, critical current density, critical fields and vortex phase diagram in pnictide FeAs-1111 and Fe-122 superconductors. It is found the anisotropy determined by the ratio of Hc2(ab, T=0)/ Hc2(c, T=0) is only about 5 in F-doped NdFeAsF (Tc=50 K), that in Ba0.6K0.4Fe2As2 is only 2. The angle dependence of resistivity in the magnetic field has also been measured and we found that the data can be scaled according to the anisotropic Ginzburg-Landau model yielding the same anisotropy. By using the value of the ratio
dHc2(T)/dT near Tc, the upper critical field Hc2(0) was determined and found to be about Hc2(c, T=0) 70 T, Hc2(ab, T=0) 300 T in F-doped NdFeAsF, and Hc2(c, T=0) 150 T, Hc2(ab, T=0) 300 T in Ba0.6K0.4Fe2As2. The higher upper critical field in FeAs-122 is attributed to the possible higher density of states in the hole doped samples.

In collaboration with Gang Mu, Zhaosheng Wang, Huiqian Luo, Huan Yang, Xiyu Zhu, Fei Han, Ying Jia, Bing Zeng, Bing Sheng. Cong Ren, Lei Shan

References


[8]. Gang Mu et al., Condmat/0808.2941.
Vidya Madhavan, Boston College

New Developments in Scanning Tunneling Microscopy of High Temperature Superconductors

What

The question of the pairing mechanism in the high temperature cuprates is as yet unsettled. Scanning Tunneling Spectroscopy (STS) is one of the best techniques to obtain information on bosonic modes (phonons, spin excitations etc) that couple to electrons. We present STS data on the electron doped superconductor Pr0.88LaCe0.12CuO4-δ (PLCCO). Below the superconducting transition temperature Tc in addition to the superconducting gap, STM spectra on PLCCO show features at approximately 3meV and 10 meV (gap-referenced) that can be associated with bosonic modes. A comparison of these energy scales with neutron scattering data on the same material indicates that the 3meV feature might originate from a phonon mode while the 10meV feature may be associated with a spin resonance mode.

We will also briefly discuss recent atomic resolution images and spectroscopy on the parent compound of a pnictide superconductor SrFe2As2. We will compare our data with LEED on identical samples and discuss the implications.
With rapid development in the past decade, ARPES has become a powerful tool to study the low energy excitations of solids, especially those novel quantum materials in which many-body physics are at play. I will first benchmark the capability of state-of-the-art ARPES system, which can now measure the Fermi surface topology to an accuracy comparable to the bulk-sensitive de Haas-van Alphen (dHvA) measurements [1,2]. With this capability, much new information on the low energy excitation in correlated electron system can be revealed. In particular, we will present our recent ARPES measurements on underdoped high-Tc superconducting cuprates. Fermi surface topology and the two-gap issue (superconducting gap vs. pseudogap) [3,4] will be discussed.


Preparing and measuring quantum states of a mechanical structure

We are performing experiments with a mechanical resonator parametrically coupled to an electrical resonator with the goal of preparing and measuring the quantum ground state and squeezed states of motion. The mechanical resonator is a very low dissipation (Q≈1M), 5 MHz, nanomechanical structure; the electrical resonator is a lithographic, low dissipation (Q≈10,000), superconducting Al, 5 GHz resonator. We pump this structure with carefully prepared microwave photons and demonstrate cooling of the mechanical structure of quantum occupation N=400 to N=20. The quantum ground state, N<1, appears within reach with a modified device (Nb, higher Q and characteristic impedance.) When we pump with two phase coherent tones, we demonstrate the back-action evading detection of a single mechanical quadrature with sensitivity of 4 times the mechanical zero-point motion. Our current measurement strength is such that we expect squeezed mechanical state to be formed. Detection with sensitivity below the zero-point level is possible in principle and appears within reach (10 GHz resonator).
An experimental search for short-length-scale deviations from Newtonian gravity (with remarks on simulating quantum magnetism using cold atoms)

This talk will describe the design of, and first results from, a new cryogenic microcantilever-based experiment for detecting deviations from Newtonian gravity at short length scales. The experiment directly measures the force between two micromachined masses separated by tens of microns. I will present the first data from the experiment, and discuss the prospects of more precisely constraining or detecting non-Newtonian effects using this probe. At the risk of cognitive dissonance, I will then very briefly describe our current work at MIT on simulating quantum magnetism using ultracold atoms in optical lattices.
Detection of weak magnetic fields with nanoscale spatial resolution is an outstanding problem in the biological and physical sciences. For example, at a distance of 10 nm, the spin of a single electron produces a magnetic field of about 1 micro Tesla, and the corresponding field from a single proton is a few nano Tesla. A sensor able to detect such magnetic fields with nanometer spatial resolution would enable powerful applications, ranging from the detection of magnetic resonance signals from individual electrons or nuclear spins in complex biological molecules to readout of classical or quantum bits of information encoded in an electron or nuclear spin memory. Here we experimentally demonstrate an approach to such nanoscale magnetic sensing, using coherent manipulation of an individual electronic spin qubit associated with a nitrogen-vacancy impurity in diamond at room temperature. Using an ultra-pure diamond sample, we achieve detection of 3 nT magnetic fields at kilohertz frequencies after 100 s of averaging. In addition, we demonstrate a sensitivity of 0.5 mT/Hz$^{1/2}$ for a diamond nanocrystal with a diameter of 30 nm.
We live in a time of extraordinary scientific, medical, and technological progress. When we look at what makes this progress possible we find a common enabling theme: mankind's ability to build ever smaller structures. How far can this era of miniaturization take us and what will be the consequences? We will explore this question by examining the ultimate limit of miniaturization: building things atom-by-atom at the nanometer scale. With some luck (keep your fingers crossed) we will connect over the internet to our laboratory in California to demonstrate how we manipulate atoms and explore what has become one of the most exciting areas of science... *The Small Frontier*
Andrea Cavalleri, Oxford UK

Ultrafast control in complex solids

What
Andrea Cavalleri, Oxford UK

Ultrafast control in complex solids

When
Thu Jan 15 8:15am – Thu Jan 15 9:15am

Created By
Aspen Winter Conference on Unifying Themes in Condensed Matter

Description
In this talk I will introduce the basic concepts of stimulated phase conversion in strongly-correlated electron systems. I will also cover past and current work of our group, which aims at designing new ways to excite a solid on the ultrafast timescale (sub-10 fs photo-doping, THz vibrational excitation, GHz pressure waves) as well as new ways to probe the system (Ultrafast X-rays).

Add to my calendar

Guests
The guest list has been hidden at organizer’s request.
James Freericks, Georgetown

Theory of time-resolved pump-probe phot... http://www.google.com/calendar/event?eid=bXM0dTEyZGJ1ZGRoZm...
Albeit most of the experiments are performed in equilibrium conditions, new horizons of condensed matter physics can be explored when a strong perturbation drives the system into a highly exited state. After the absorption of an ultrashort laser pulse, the interatomic forces that bind the atoms can be substantially altered, leading to propagation of lattice waves or to sudden structural changes. Since the pathway to non equilibrium states evolves on the femtosecond timescale, any experimental observation of the nuclei and electrons motion is especially challenging. In our approach, an intense and ultrashort laser pulse perturbs the system, while an ultraviolet source induces the emission of photoelectrons after a variable time delay. Such technique exploits the high potentials of photoelectron spectroscopy for the investigation serveral interesting topics. We will discuss ultrafast insulator-metal transitions in Mott insulators, coherent lattice motion in charge density wave materials, and electron-phonon coupling in high temperature superconductors. Our results show that time resolved techniques provide novel information on many-body interactions that would not be accessible by standard means.
Antti-Pekka Jauho, TU Denmark  

**Thermal rectification in nonlinear quantum circuits**

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<th>Antti-Pekka Jauho, TU Denmark</th>
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<td>Description</td>
<td>We present a theoretical study of radiative heat transport in nonlinear solid-state quantum circuits. We give a detailed account of heat rectification effects, i.e. the asymmetry of heat current with respect to a reversal of the thermal gradient, in a system consisting of two reservoirs at finite temperatures coupled through a nonlinear resonator. The inclusion of nonlinear effects generalizes our recent work on exactly solvable linear systems [1]. We suggest an experimentally feasible superconducting circuit employing the Josephson nonlinearity to realize a controllable low temperature heat rectifier with a maximal asymmetry of the order of 10%. Strikingly, we discover that rectification can change sign as a function of temperature. (*) Work done in collaboration with T. Ruokola and T. Ojanen</td>
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Add to my calendar
**What**  
Hidemi Shigekawa, Tsukuba, Japan  
*Imaging carrier dynamics in nanoscale potential landscapes by laser-combined scanning tunneling microscopy*

**When**  
Thu Jan 15 5:15pm – Thu Jan 15 5:45pm

**Created By**  
Aspen Winter Conference on Unifying Themes in Condensed Matter

**Description**  
Since the invention of scanning tunneling microscopy (STM), direct imaging of atomic-scale structures has been raising the veil from various longstanding problems in and developing frontiers of science and technology. However, since the temporal resolution of STM is limited to less than 100 kHz, the transient carrier dynamics in materials, the understanding and control of which is the essential basis for the future advancement of nanoscale science and technology, has been beyond its field of vision. Here, we introduce a new STM-based microscopy technique, which enables us to visualize the dynamics of the excited carriers modulated in nanoscale potential landscapes. Ultimate spatial and temporal resolutions were simultaneously realized by combining advanced laser technology with STM.


Exciton Diffusion Measurements in Organic Semiconductors

Organic semiconductors combine novel semiconducting optoelectronic properties with the scope for simple fabrication and tuning of properties. An important class of these materials are conjugated polymers which are model one-dimensional systems in which electrons correlations play a prominent role, leading to strongly bound excitons as the primary photoexcitations. The development of solar cells from these materials requires these excitons to be dissociated which can be achieved by them diffusing to a junction with another material (of higher electron affinity). The distance an exciton can diffuse is therefore a key issue in the operation of these solar cells. We have therefore developed ultrafast spectroscopy techniques for studying exciton diffusion. by measuring processes such as exciton-exciton annihilation, transient fluorescence in the presence of a quencher, and fluorescence anisotropy, the exciton diffusivity can be deduced. These techniques open up a new field of exciton transport in organic semiconductors.

What happens when nanostructures are pushed to the atomic limit? This talk addresses some of the consequences, such as fabricating atomically-precise nanostructures by self-assembly [1], exploring the behavior of one-dimensional electrons traveling along an atom chain [2], measuring the wave functions of electrons in nano-objects, and finding fundamental limits of data storage [3].

Semiconductor surfaces lend themselves towards self-assembly, because the broken bonds create elaborate reconstruction patterns to minimize the surface energy. The surface electrons at the Fermi level de-couple from the substrate, because their energy lies in the band gap. One-dimensional atom chains at silicon surfaces display surprising features [2], such as fractional band filling, a spin-splitting of the Fermi surface obtained from non-magnetic atoms, the formation of graphitic ribbons of Si hexagons, and nano-scale phase separation of a chain into metallic and semiconducting sections. For fully understanding these exotic phenomena one would want to measure wave functions of electrons in a nanostructure, either in real space by scanning tunneling spectroscopy or in reciprocal space by momentum-resolved photoemission. Are these two techniques measuring the same quantity? Can they be connected by a generalized Fourier transform?

An array of self-assembled atomic chains can be used as patterned data storage medium which stores a bit by the presence or absence of a single silicon atom. Such a toy memory is used to explore fundamental limits of data storage and to compare silicon with DNA as media for information storage [3].


Jan von Delft, LMU Munich
Kondo decoherence: finding the right spin model for iron impurities in gold and silver

We exploit the decoherence of electrons due to magnetic impurities, studied via weak localization, to resolve a longstanding question concerning the classic Kondo systems of Fe impurities in the noble metals gold and silver: which Kondo-type model yields a realistic description of the relevant multiple bands, spin and orbital degrees of freedom? Previous studies suggest a fully screened spin S Kondo model, but the value of S remained ambiguous. We have performed density functional theory calculations that suggest S = 3/2. We also compare previous and new measurements of both the resistivity and decoherence rate in quasi 1-dimensional wires to numerical renormalization group predictions for S=1/2, 1 and 3/2, finding excellent agreement for S=3/2.

We use scanning micro-SQUID susceptometers to study persistent currents in superconducting and normal metal rings. In superconducting Al rings, we find (contrary to previous results) that the measured susceptibility as a function of applied field and temperature agrees quantitatively with a Ginzburg-Landau-based model of thermal fluctuations. This result is particularly interesting in the Little-Parks regime near a half-quantum of applied flux where superconductivity is destroyed, and demonstrates the feasibility of measurements in the so-called destructive Little-Parks regime near zero temperature, where applied magnetic flux causes a series of superconducting-normal and normal-superconducting transitions and quantum fluctuations are expected to be very important. In normal-metal gold rings, we find an $h/e$-periodic susceptibility in sufficiently small rings, a signature of persistent currents: again contrary to previous results, we find that the susceptibility agrees quantitatively with what is expected for a normal-metal ring in the diffusive limit. If time permits, I will also discuss our observation on various metallic and insulating films of a linear paramagnetic signal, indicating the presence of a population unpaired defect spins, accompanied by a poorly understood zero-field anomaly.
Upon increasing the density of electrons in a quantum wire, the system undergoes a transition from a one-dimensional to a quasi-one-dimensional state. We study the evolution of the system and the electronic excitation modes in the vicinity of the transition as a function of the interaction strength. In particular, we establish that, for spin-polarized electrons, only one gapless mode exists on either side of the transition at any interaction strength [1]. In the strongly interacting regime, the effective Hamiltonian is represented by two weakly coupled modes given by a Luttinger liquid and a transverse field Ising model. Performing a renormalization group analysis, we show that the critical fixed point is Lorentz invariant. However, the critical velocity vanishes due to marginally irrelevant operators [2].


Recent progress on scanning tunneling microscopy (STM) has made it possible to position atoms and probe spin excitations in atomic-scale accuracy on surfaces, including spin coupling between atoms [1] and magnetic anisotropy of a single atom [2]. We have determined the precise local structure around the surface impurities using density functional theory (DFT). From that we find the magnetic atoms can form molecular structures with its binding surface, behaving like surface molecular magnets. The charge transfers between the magnetic atom and its neighboring atoms are also calculated to determine the bond nature. In studying spin coupling, we apply GGA+U to calculate the exchange coupling J (several meV) for a Mn dimer on the surface. The calculated J’s reproduce the dimer’s antiferromagnetism obtained from the STM experiments, and are within 8% of the experimental values. To demonstrate the potential to engineer the coupling between atomic spins, we calculate the J’s for the Mn dimers atop different sites on the surface, and find the J values can be different by twice. Although ferromagnetic coupling of engineered atomic spins has not been observed, we have calculated J of a Gd dimer on the same surface and find that it exhibits a weak ferromagnetism. Such a finding has demonstrated a great potential in building an atomic-sized magnet that can be engineered. In studying magnetic anisotropy of a single atom, we calculate and investigate the three-dimensional spin density distribution of a single Fe on a surface. We find the most anisotropic orientation of the spin density distribution is consistent with the primary axis of the system’s magnetocrystalline anisotropy measured by STM. We also find significant spreading of spin density from the Fe binding site, which is similar to that reported in DFT calculations of molecular magnets. As an ongoing search for larger magnetic anisotropy of similar systems, initial results of magnetic anisotropy of Tb on the same surface will also be presented.

Science 312, 1021 (2006).