

7. HIGHLIGHTS

Facilities

Laboratory for Integrated Science and Engineering & Center for Nanoscale Systems
— Harvard University

National Nanostructure Infrastructure Network (NNIN) — Harvard University and
the University of California at Santa Barbara

Education and Outreach

NISE Network

New Internship Program

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— **George M. Whitesides**

CMOS Mini NMR System — **Donhee Ham**

Electroactive Hydrogels for Biological Applications — **Kevin (Kit) Parker**

Size Segregation of Giant Unilamellar Vesicles — **Howard A. Stone**

Fluorescent and Raman Active Silver Nanoparticles — **Xiaowei Zhuang**

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Laboratory for Integrated Science and Engineering & Center for Nanoscale Systems Harvard University



Goals

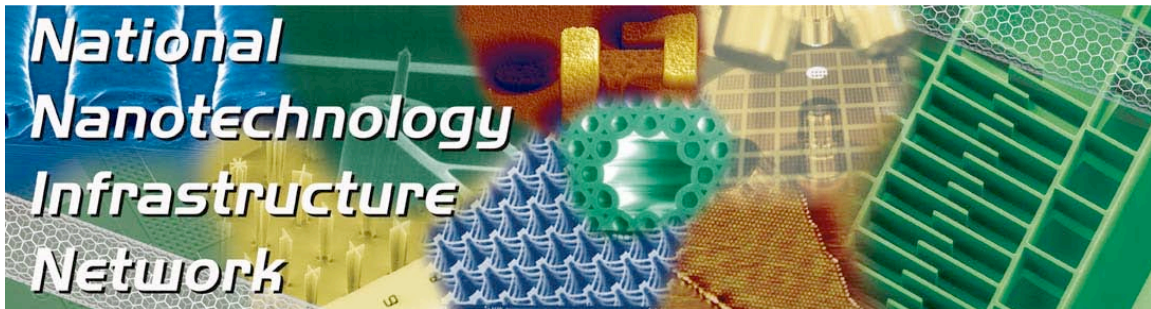
To provide world-class, centralized facilities and technical support for Harvard faculty research groups as well as the larger community of external users from academia and industry.

To foster leading-edge, multi-disciplinary research and education in the area of imaging and nanoscale systems, bridging the disciplines of chemistry, physics, engineering, materials science, geology, biology, and medicine.

To create an environment for collaborative research by providing shared research facilities and meeting places conducive to productive scientific interactions.

National Nanostructure Infrastructure Network (NNIN)

Harvard University and
University of California at Santa Barbara



Harvard and UC Santa Barbara are two of an integrated partnership of thirteen user facilities led by Cornell and Stanford that provide opportunities for nanoscience and nanotechnology research. At Harvard, the NNIN provides expertise in computation and in soft lithography and assembly through the Center for Nanoscale Systems. At UCSB, the NNIN provides expertise in optics and electronic materials. The NNIN was funded by the NSF in January 2004.

Nanoscale Informal Science Education NISE Network

Carol Lynn Alpert, Larry Bell, Kathryn Hollar, Eric Mazur, Robert Westervelt, George Whitesides



(Left): A young audience member assists grad student Tom Hunt demonstrate atomic force microscopy at the Museum of Science, Boston.
(Right): Michael Stopa discusses privacy issues during a NanoFutures Forum at the Museum of Science.

Our NSEC is a strong partner in the Nanoscale Informal Science Education (NISE) Network that links science museums, educational organizations and research institutions. The Center's faculty and students, guided by our Education Director Kathryn Hollar, develop interactive demonstrations and help set up NanoFutures Forums at the Museum of Science, Boston, as well as participate in the NISE Nanoscale Education Outreach (NEO) Workshops at the Exploratorium in San Francisco. The NISE Scientific Advisory Board includes NSEC faculty Eric Mazur, Robert M. Westervelt, and George M. Whitesides.

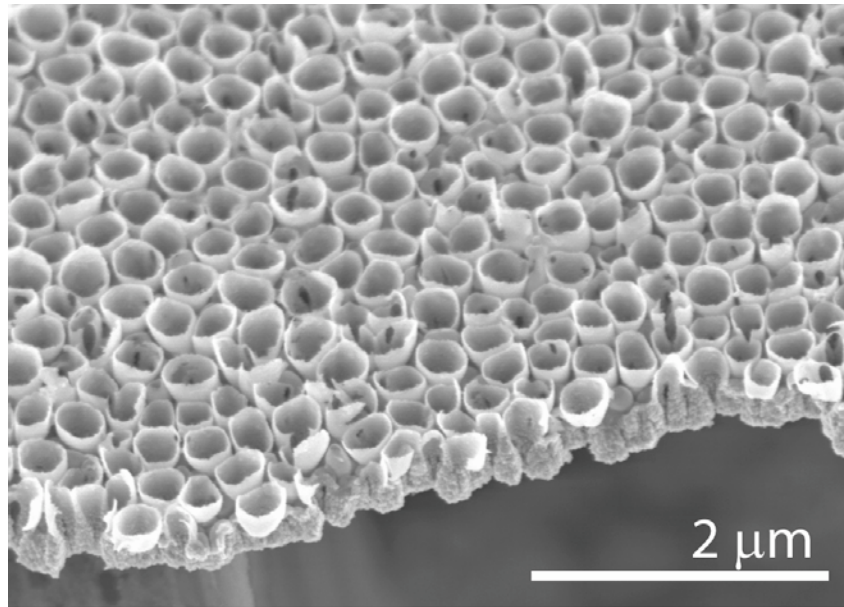
New Internship Program Museum of Science, Boston



In 2007, with the full support of Robert M. Westervelt, the Museum of Science pioneered a new program that brought NSEC graduate students from the NSEC into the museum for an intensive one-week informal science communication training internship. This practicum culminated in the development of live public presentations given by the students. Here Tina Shih (*at right*) speaks with a group of visitors after the conclusion of her presentation. The internship program is intended to help early career researchers appreciate the importance of engaging public audiences in current research, and to help them develop skills in this area. The training can also help students communicate to fellow researchers across disciplines, a necessity in nanoscale science and engineering.

Fabrication of Arrays of Metal and Metal-Oxide Nanotubes by Shadow Evaporation

George M. Whitesides

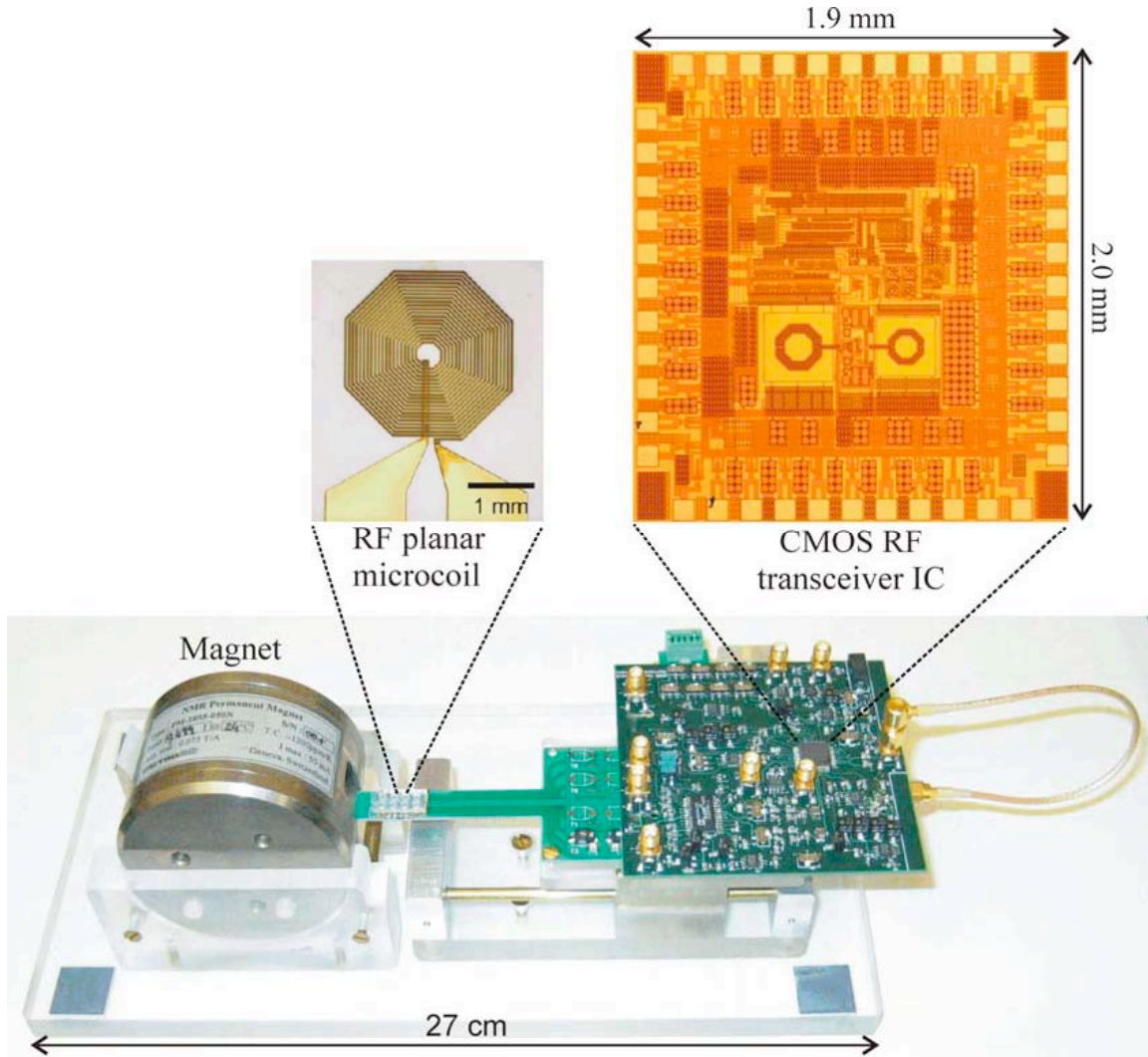


A SEM image of indium-tin oxide (ITO) nanostructures formed by line-of-sight evaporation into sacrificial anodized aluminum oxide pores. The heights and diameters of the tubes are ~ 200 nm.

We developed a simple technique that involves depositing material onto an anodized aluminum oxide (AAO) membrane template using a collimated electron beam (e-beam) evaporation source. The evaporating material enters the porous openings of the AAO membrane and deposits on to the walls of the pores. The membrane is tilted with respect to the column of evaporating material, so the shadows cast by the openings of the pores onto the inside walls of the pores defines the geometry of the tubes. Rotation of the membrane during evaporation ensures uniform deposition inside the pores. After evaporation, dissolution of the AAO in base easily removes the template to yield an array of nanotubes connected by a thin backing of the same metal or metal-oxide. The diameter of the pores dictates the diameter of the tubes, and the incident angle of evaporation determines the height of the tubes.

CMOS Mini NMR System

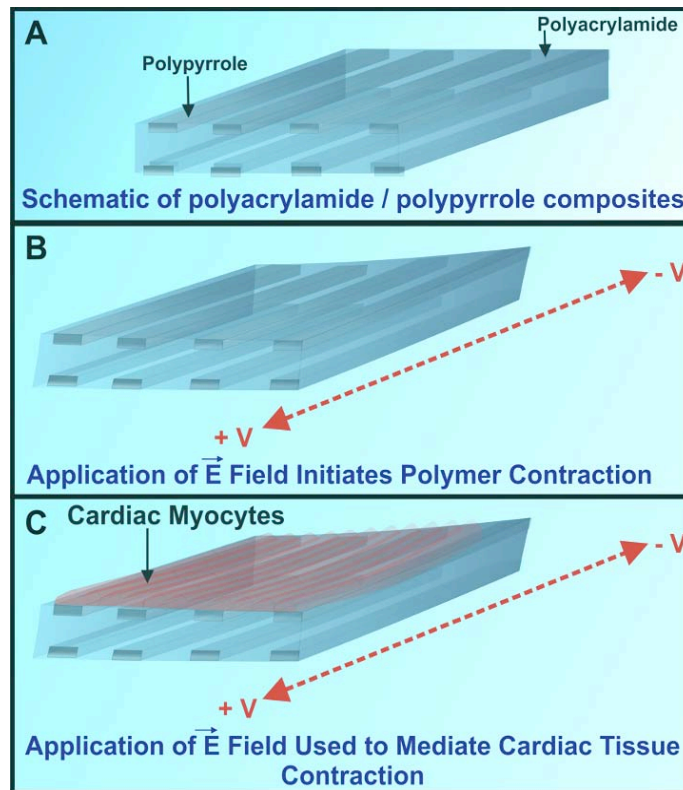
Donhee Ham



Donhee Ham's group at Harvard, in collaboration with Ralph Weissleder's group at MGH, has developed the smallest complete NMR relaxometry system (Figure above), which is 40 times smaller, 60 times lighter, yet 60 times more sensitive than a commonly-used, state-of-the-art commercial benchtop system. The key to this development is the CMOS integration of a highly sensitive and versatile RF transceiver, that can drive and monitor the nuclei spins in samples. This system can detect biomolecules (e.g., cancer protein markers) by monitoring nuclear spin relaxation times, in conjunction with magnetic nanoparticles, and it can be used as a portable diagnostic tool when it is fully developed.

Electroactive Hydrogels for Biological Applications

Kevin (Kit) Parker



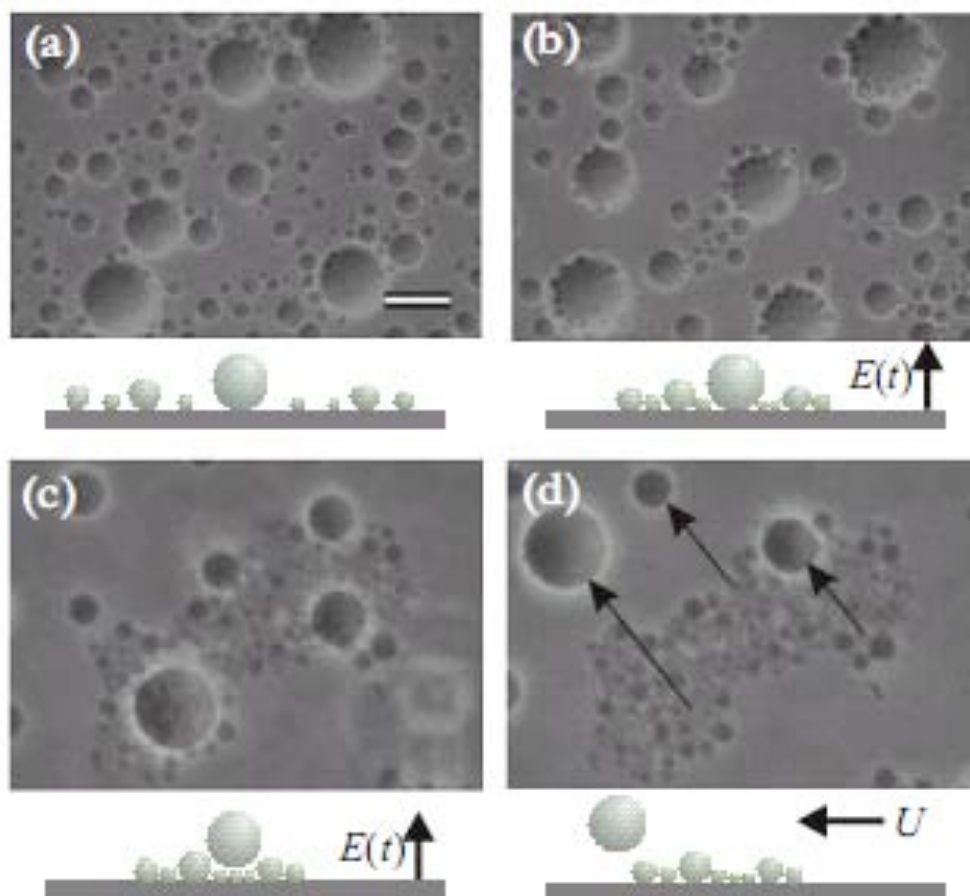
We have constructed thin ($\sim 25 \mu\text{m}$) PAA/PPy hydrogels and are testing their generation of forces vs. applied voltage. (A) Design of a polyacrylamide/polyppyrrrole hydrogel which deforms in the presence of an electric field. (B) Nanoscale patterning of polyppyrrrole allows for precise control of hydrogel deformation by an electric field. (C) The electroactive hydrogels can be coupled with live cells to dynamically control the mechanical properties of a tissue/polymer biohybrid.

Electroactuated polymer hydrogels are attractive materials for biological applications, namely because they can operate in physiological solutions, require low voltages for actuation, are biocompatible, and can be easily microfabricated. Although electroactive hydrogels have been developed for artificial muscle applications, the ability to control stress gradients in a hydrogel via the application of an external electric field has not been demonstrated.

Megan O'Grady and Kevin (Kit) Parker, Disease Biophysics Group, Harvard University

Size Segregation of Giant Unilamellar Vesicles

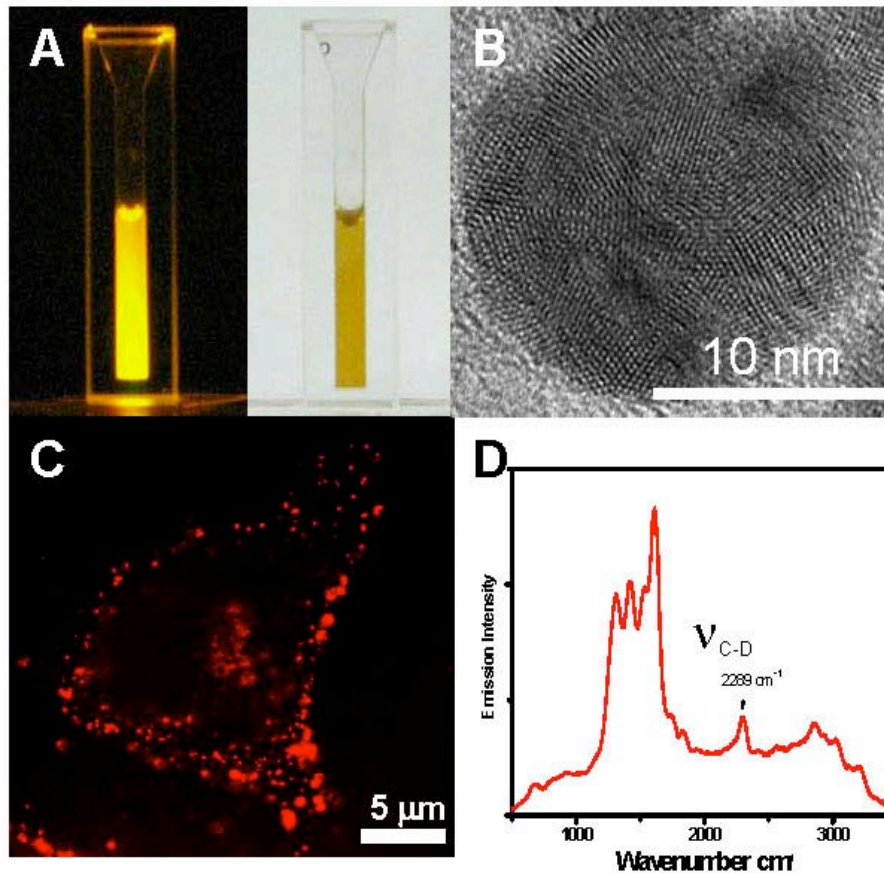
Howard Stone



We have developed an electro-hydrodynamic approach that separates giant unilamellar vesicles (tens of microns in diameter) from a suspension with vesicles from submicron to tens of microns in diameter. **(a)** Vesicles arranged randomly near the electrode. **(b)** 30 seconds after application of a 10 V, 30 Hz AC field, smaller vesicles aggregate around larger ones. **(c)** After 10 minutes of the electric field, larger vesicles have moved above smaller vesicles. **(d)** After a short DC pulse adheres smaller vesicles to the electrode, a pressure-driven flow (in direction of arrows) removes the larger vesicles. Scale bar in **(a)** is 25 μm .

Fluorescent and Raman Active Silver Nanoparticles

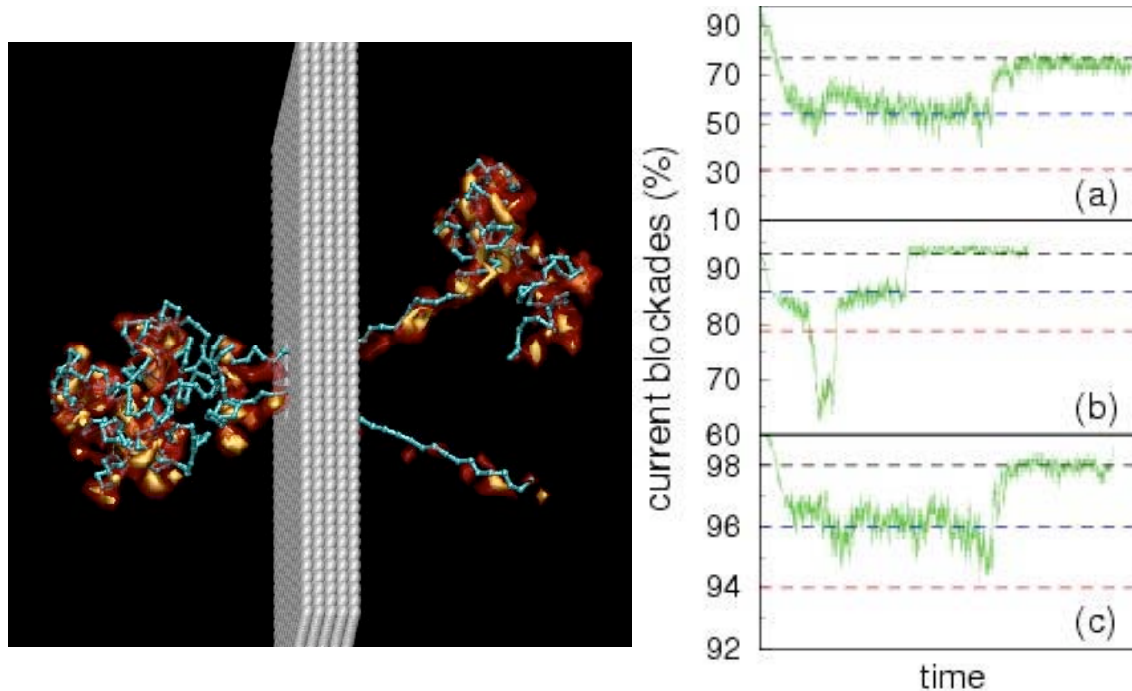
Xiaowei Zhuang



(a) Photos of the silver nanoparticle solution with (left) and without (right) laser excitation. **(b)** High resolution TEM image of a single silver nanoparticle. **(c)** A live HeLa cell labeled with Fluorescent and Raman active silver nanoparticles. **(d)** A surface-enhanced Raman spectrum of a single silver nanoparticle on the live HeLa cell surface in 90 $\mu\text{g/ml}$ deuterated-glycine solution. Stretch vibration of C-D of deuterated-glycine was clearly detected by the silver nanoparticle.

Multiscale Modeling of DNA Translocation through Nanopores

Efthimios Kaxiras

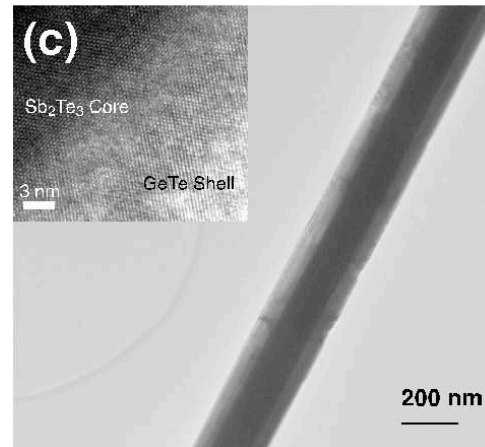
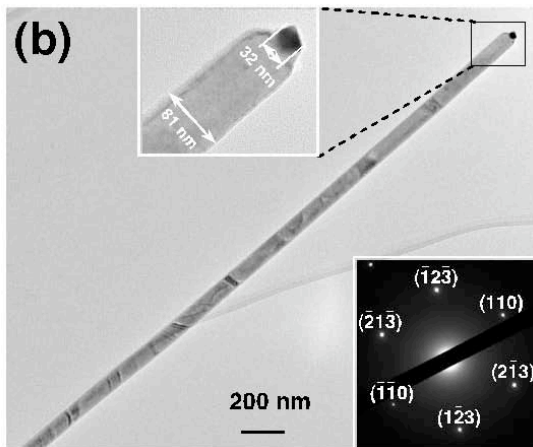
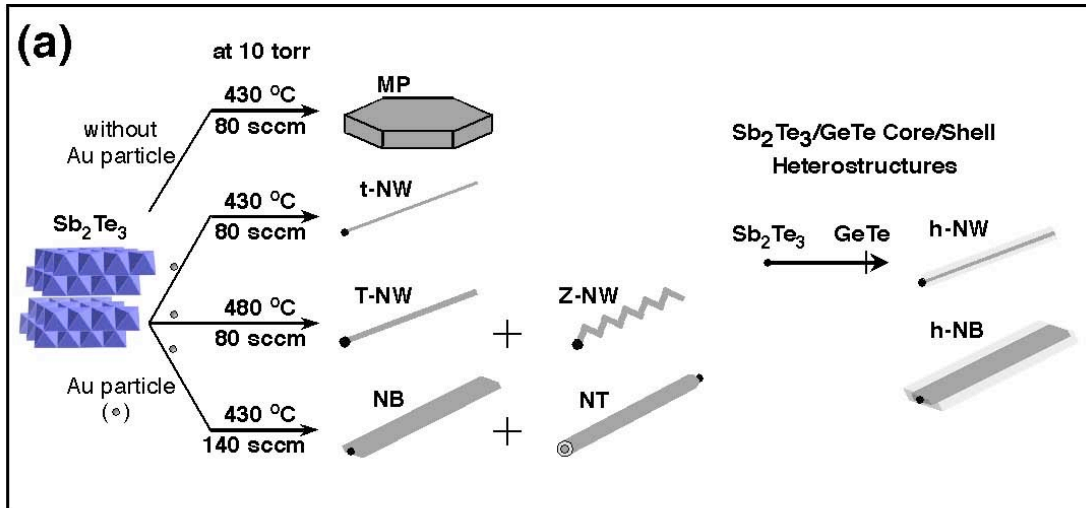


Left panel: 3-D visualization of the translocation process of DNA passing through a nanopore: The molecule is shown as cyan beads, and the fluid velocity by 3-D contours in the vicinity of the beads. **Right panel:** Current blockades vs. time for (a) small, (b) mid-sized and (c) large pore diameters. Red, blue and black dashed lines correspond to 3-fold, 2-fold and single-fold biopolymer conformations translocating through the pore.

DNA sequencing by solid nanopores promises much greater speed than existing methods and could change the nature of medical diagnosis. To understand the physics, we use an innovative multi-scale methodology, which treats a biopolymer (like DNA) translocating through a nanopore in the presence of a solvent. Our work provides evidence for quantized ion-current blockade and its direct connection to the folding configuration of the biopolymer, as well as detailed information on the role of the hydrodynamic correlations in speeding-up the translocation process. The acceleration is interpreted as an interplay between the pore geometry and the fluid motion. The insight from these simulations suggests prospects for optimized nano-hydrodynamic devices, like multi-translocation chips, based on the fine-tuning of pore structure and hydrodynamic correlations.

Phase-Change Nanowires and Nanowire Heterostructures

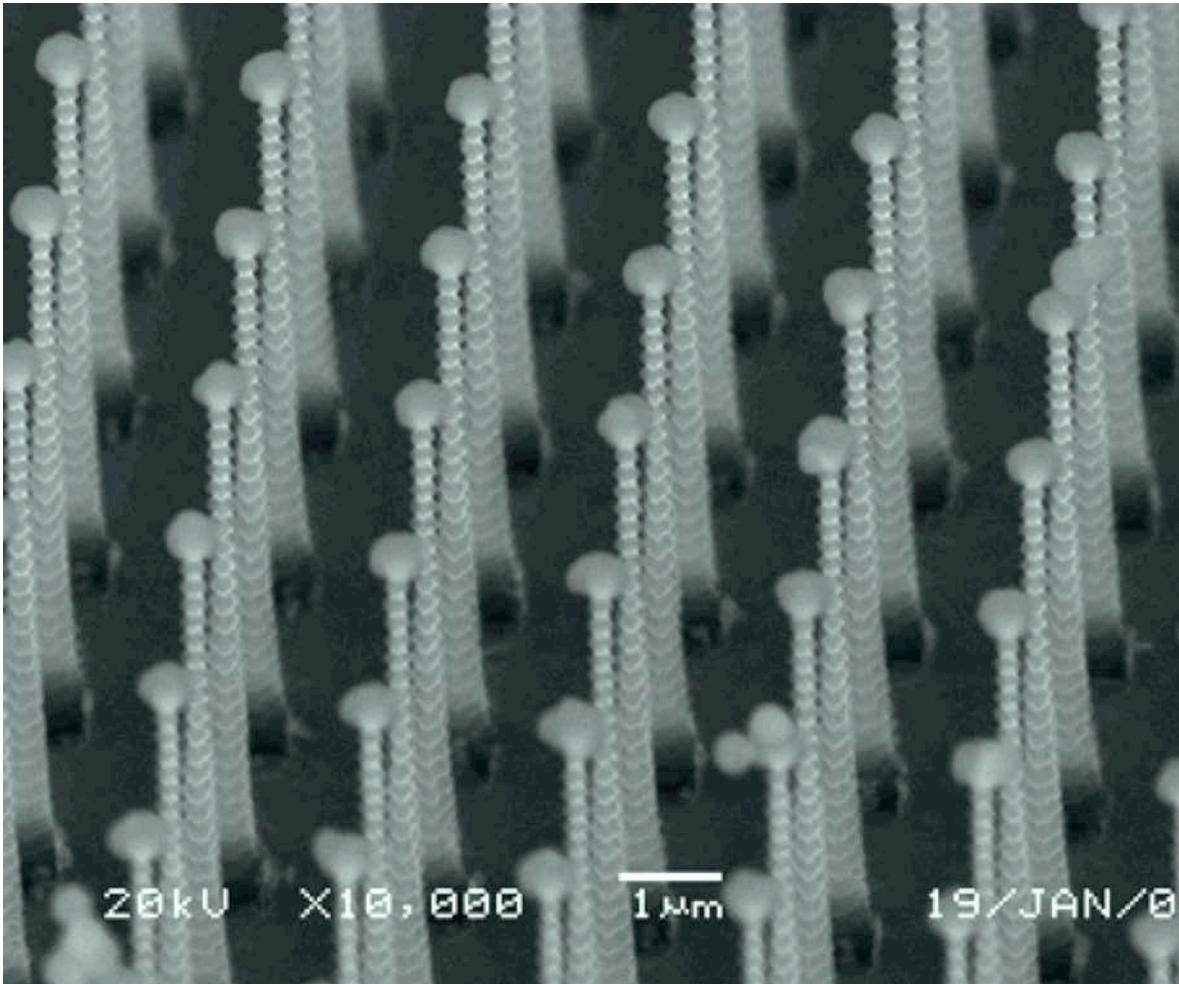
Hongkun Park



Chalcogenides exhibit a reversible crystalline-amorphous phase change induced by temperature or electric field, and form the material bases for data storage media such as CD/DVD and phase-change random access memory (PRAM). The intrinsic properties of chalcogenide materials and their size-dependent variations determine the speed, stability, and miniaturization of PRAM, highlighting the importance of studying these phase-change materials in a nanostructured form. In 2007 Park developed a new method to synthesize single-crystalline phase change nanowires and nanowire heterostructures composed of Sb_2Te_3 and GeTe.

New Method of Localized Materials Deposition on a Nanowire Array: Nanoscale Building Blocks

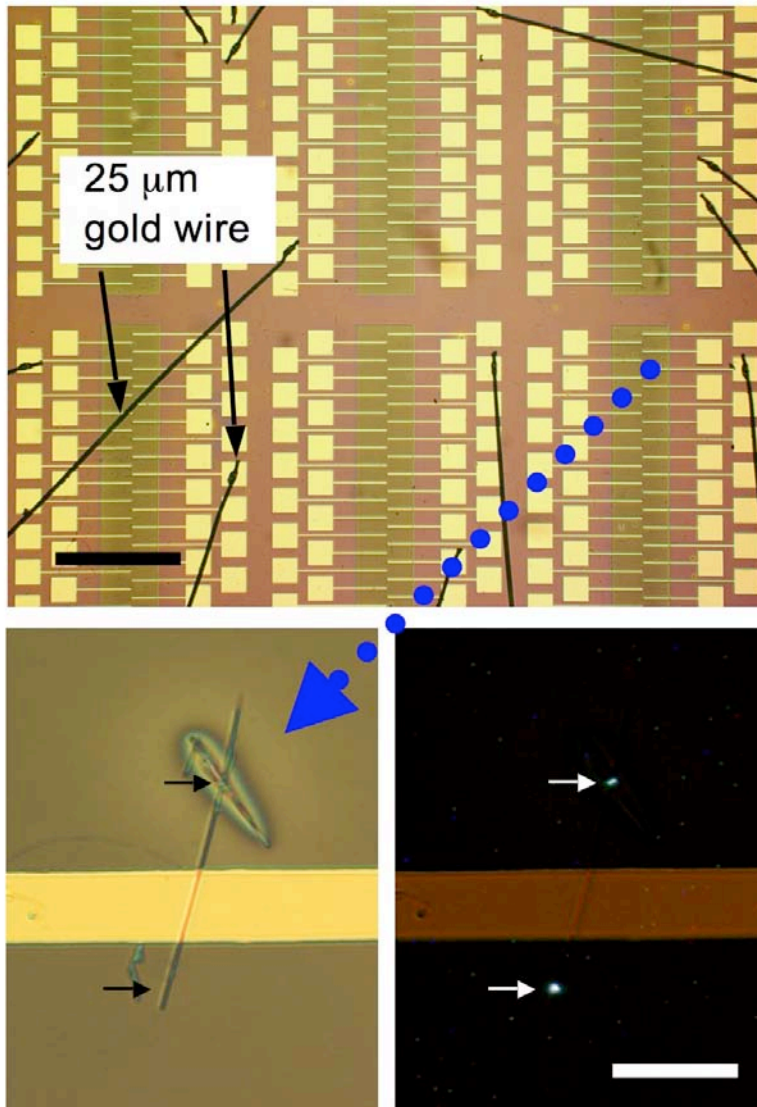
Joanna Aizenberg



We have developed a new approach for the fabrication of asymmetric nanowire arrays, in which a guest material is deposited only at one end of each nanowire due to the super-hydrophobic character of the nano-structured surface. Highly uniform deposition of particles with controlled size, geometry and structure occurs, as shown for the calcium carbonate particles shown above. The method could be applied quite generally to a wide range of nucleation/growth precipitation reactions producing novel functionalized asymmetric nanowire arrays and 3-D nanostructures for applications in electronic and optical materials, actuators, biological sensors, functional “barcode” designs, and self-organization.

Nanowire Optoelectronics

Federico Capasso and Lars Samuelson

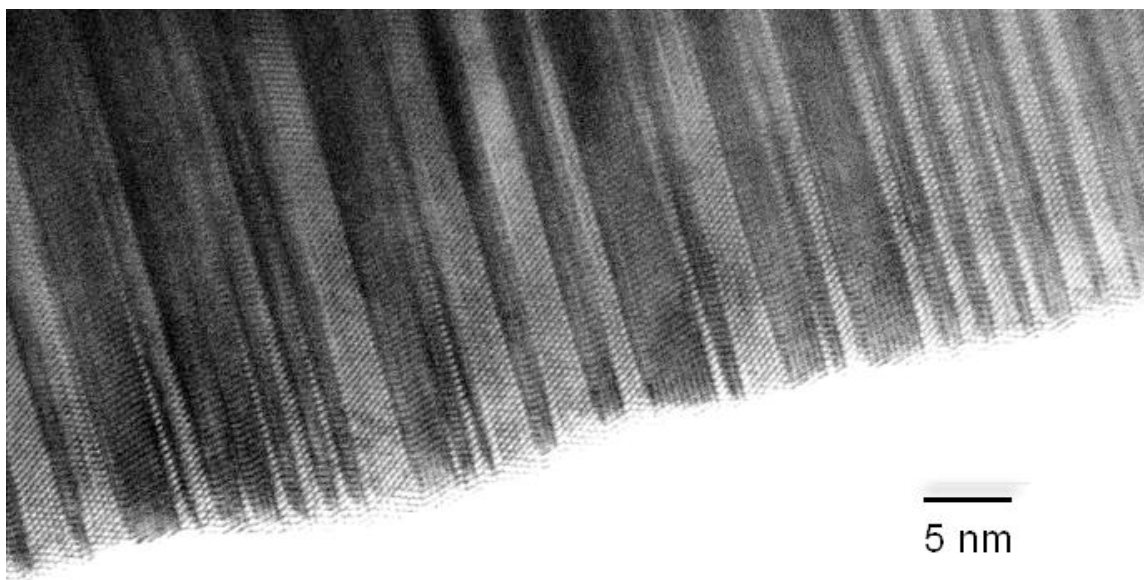


We have developed a new method to fabricate single-nanowire light-emitting diodes (LEDs) over a large area using a combination of spin-on glass technology and optical lithography. This method provides a powerful, fast, and inexpensive way to characterize nanowire electronic and photonic devices. Also, in combination with a nanowire alignment technique, it could form the basis of future integrated photonic circuits, where light rather than electrical current carries the

information. The figure (**top**) shows an optical micrograph of a finished wafer containing hundreds of ultraviolet single-nanowire LEDs, using zinc oxide nanowires. The scale bar is 1 mm. The bottom figures are zoomed-in images of a single device under an applied voltage, emitting ultraviolet light at room temperature (as indicated by the arrows). The scale bar is 30 μm .

New Nanowire Heterostructures

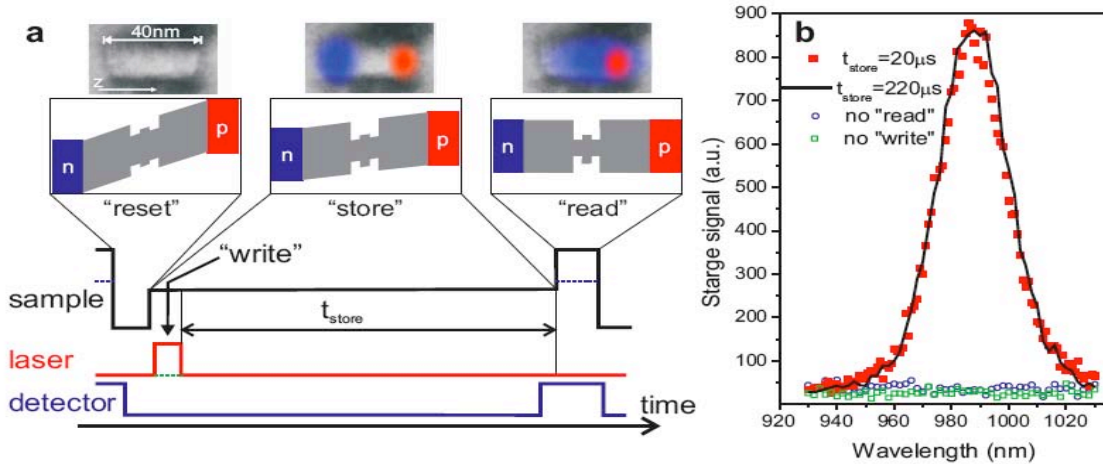
Federico Capasso and Lars Samuelson



We have discovered a new type of heterostructure, which forms at the nanoscale from the alternation of different crystal orientations within the *same* chemically homogeneous semiconductor. The figure shows a high-resolution transmission electron microscope image of such a material (an InP nanowire), where the different layers can be clearly seen. Each alternate section has the same crystal structure but it is rotated by 180 degrees from the adjacent layers. This is called rotation twinning. We have deduced the optical and electronic properties of this novel heterostructure from photoluminescence measurements combined with quantum mechanical calculation of the electron and hole states. This discovery opens up new opportunities in material engineering since it makes it possible to tune the electronic properties of the material by using twinning.

Unique Exciton Storage Memory

Pierre M. Petroff

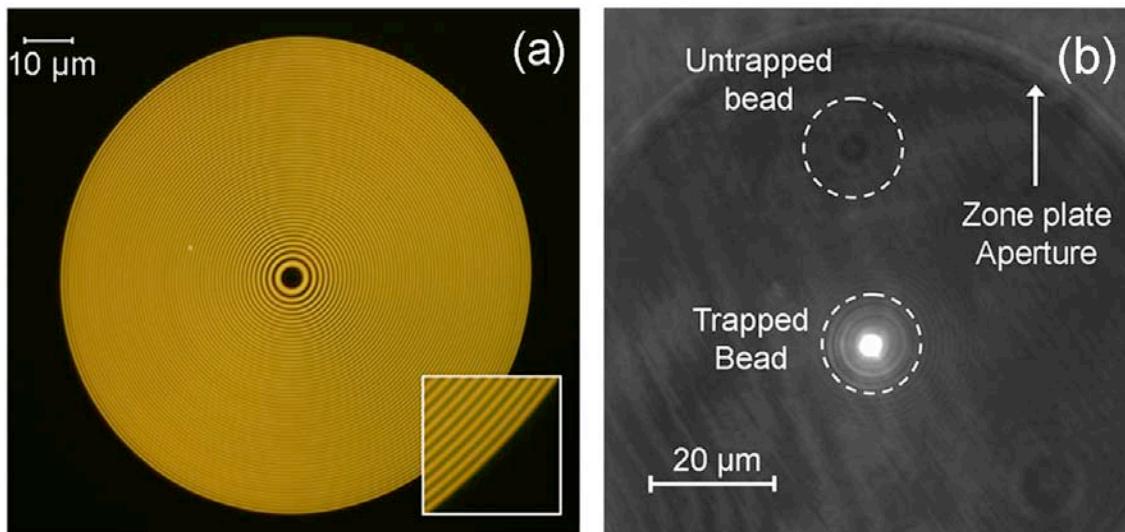


Storage of excitons in 40 nm high quantum particles (QPs). **(a)** Schematic of the storage scheme. After a reset step during which the QP is emptied the sample voltage (black) is switched to the indirect exciton regime. In the beginning of the storage interval, electrons and holes are written by a laser (red) into the QP and after t_{storage} read via direct excitons. The detection (blue) is activated only during the time of the read step. The band structure and spatial carrier distribution during each step are shown schematically. For control experiments, the write and read pulses are turned off individually as shown by the dashed lines. **(b)** Spectra taken for 20 ms (full squares) and 200 ms (line) storage times ($P_{\text{exc}} = 20 \text{ kW/cm}^{-2}$, $T = 7 \text{ K}$). Without the "write" and "read" pulses (open symbols), no signal is detected as expected.

We have developed a *unique exciton storage memory* based on a novel self-assembled nanostructure, which is composed of two quantum dots connected by a short quantum wire. Optically or electrically injected excitons can be stored for milliseconds at temperatures up to 100 K. This exciton memory, which is read optically, can function as a single photon emitter on demand and should enable a controlled single spin manipulation with application to quantum cryptography and quantum computing.

Microfabricated Fresnel Zone Plate Optical Tweezer

Kenneth B. Crozier

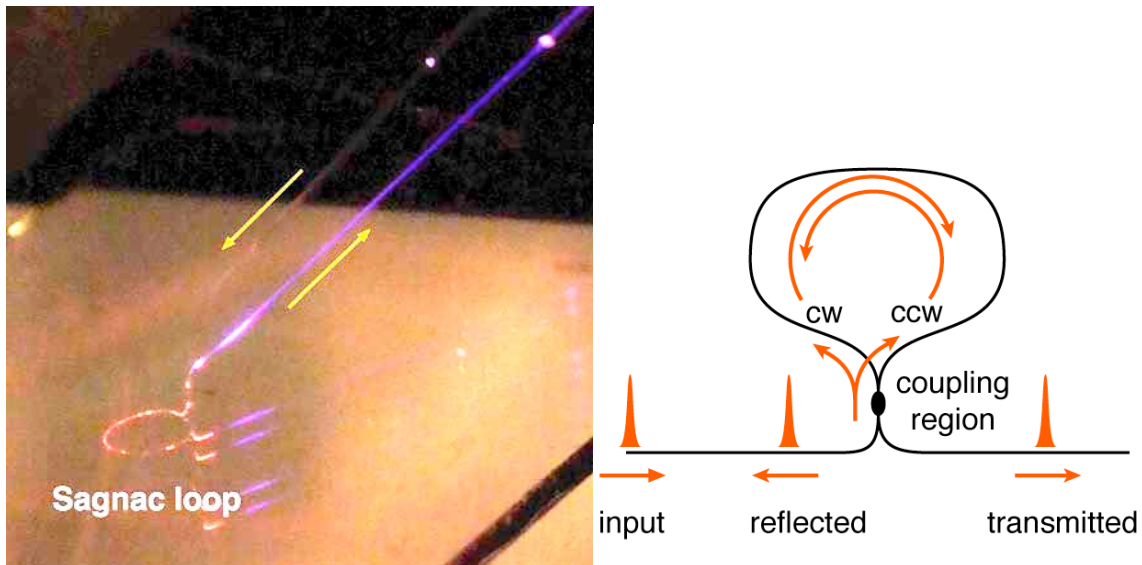


(a) Photograph of microfabricated Fresnel-Zone-Plate optical tweezer, consisting of concentric gold rings (50 nm thick) on a microscope slide. The Zone Plate outer diameter is 100 μm, and the focal length is 8 μm. **(b)** CCD camera image of a fluorescent bead (2 μm diameter) trapped in Zone Plate focus.

The incorporation of optical tweezers into microfluidic chips would provide new functionalities for these systems, such as particle sorting, particle manipulation, and biophysical force measurements. This motivates the development of microfabricated structures for optical trapping. We have demonstrated the use of a Fresnel zone plate as an optical trap. Electron-beam lithography is used to produce the device, which consists of concentric gold rings on a glass substrate **(a)**. A collimated laser beam is focused by the zone plate to a submicron spot. In **(b)**, a photograph of a 2 micron bead trapped by the Zone Plate is shown.

Nonlinear Sagnac Interferometer

Eric Mazur

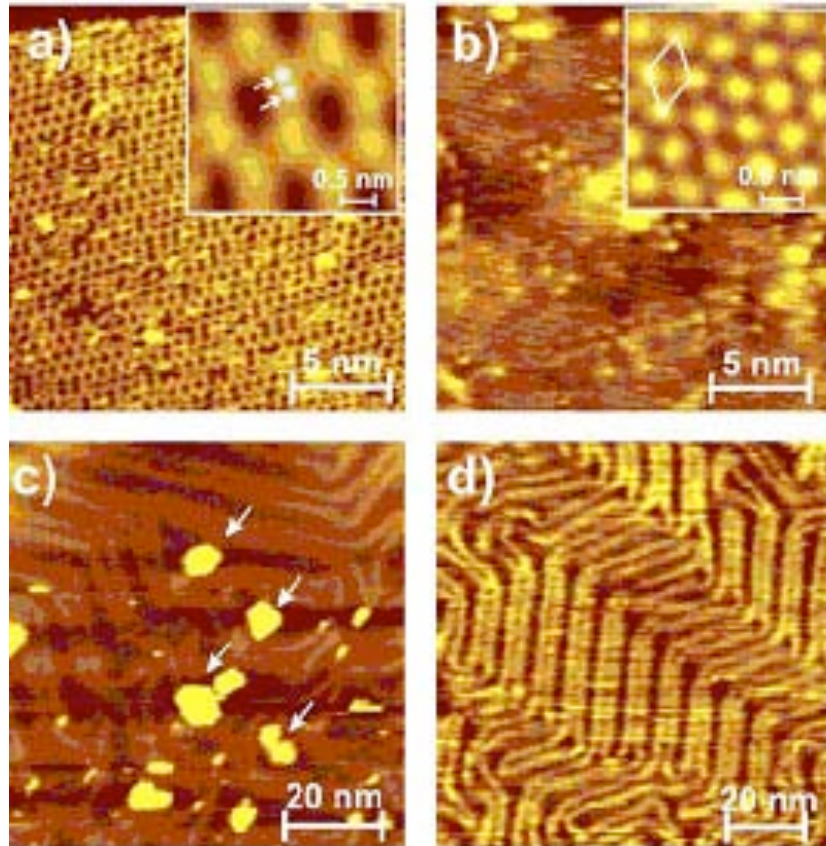


(*Left*) Photograph and (*right*) diagram of a nonlinear Sagnac interferometer showing the input pulse, the counter-propagating paths, the reflected and transmitted pulses.

Silica fibers, whose diameters are smaller than the wavelength of light, can be twisted into a loop structure that behaves as a Sagnac interferometer. When ultrashort laser pulses (~ 100 fs) are launched into the loop, the nonlinear effects create a device with a power-dependent transmission. By tuning the coupling region and the input pulse power, the Sagnac loop can perform optical switching as well as optical logic operations.

Chemical Mobilization of Metal Atoms in Nanostructures

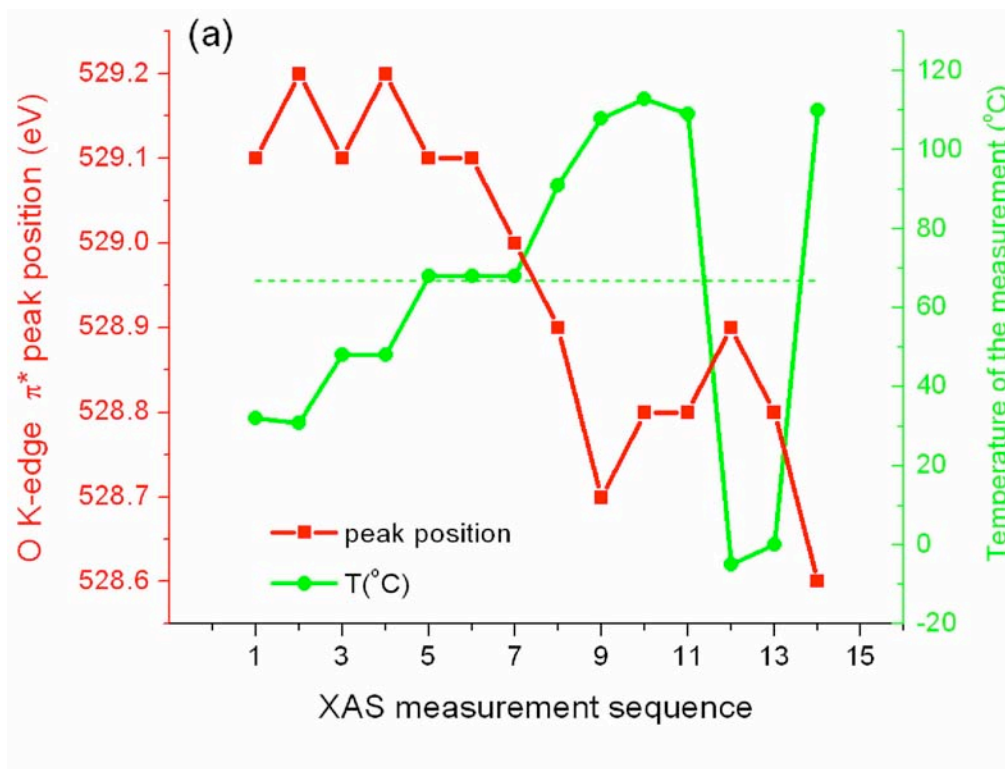
Cynthia M. Friend



Scanning tunneling microscope images of chlorine atoms on Au illustrate the importance of the chemical environment on the ability to form and retain the structure of nanostructures. Chlorine atoms form a structure that lifts Au out of the bulk at high Cl concentrations (~ 0.8 Cl atoms per Au surface atom) (*top left panel, a*). As the surface is heated, Cl leaves the surface and intermediate structures are formed that illustrates the release of Au atoms. Panel *c* (*lower left*) clearly shows nanoscopic Au islands. When all Cl is removed, the surface reverts to the characteristic “herringbone” structure (*lower right panel, d*). We have also performed theoretical calculations on these systems to develop an understanding of the energetic factors in determining the structures formed. These data are reproduced from W.W. Gao *et al.*, *J. Am. Chem. Soc.* (2008), in press.

Mapping the Electronic Structure of VO₂ Thin Films Using NEAFS across the Phase Transition Boundry

Shriram Ramanathan

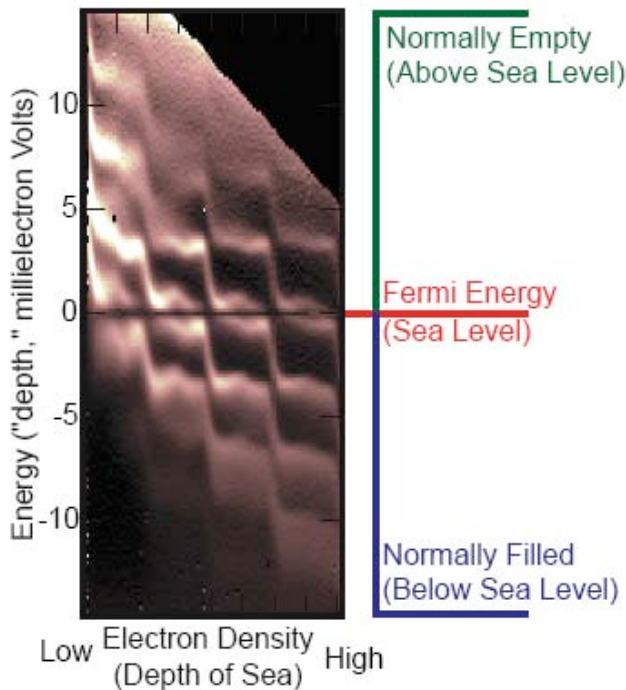


X-ray absorption spectroscopy (XAS) and X-ray photoemission spectroscopy were performed on VO₂ thin films rf sputtered at various conditions, showing the V *L*-edge and O *K*-edge. The spectra show changes in the electronic structure depending on the film quality. XAS of the O *K* edge show that the spacing between *3d-π* and *3d-σ* bands decreases by 0.8 eV with concurrent broadening of both bands for the sample sputtered at lower substrate temperature, which is more polycrystalline and disordered. The *3d-σ* band position appears to be more sensitive to the sample quality, indicating that the cation-ligand interaction is mostly affected likely due to the distortion of the local O coordination surrounding a V ion. The thermal cycling of the VO₂ films through the metal insulator transition (MIT) shows irreversible shifts of the conduction bands toward lower photon energies,

that are apparently caused by sample deterioration caused by lattice transformations at the MIT.

Spectroscopy of Quantum Hall Systems

Raymond Ashoori



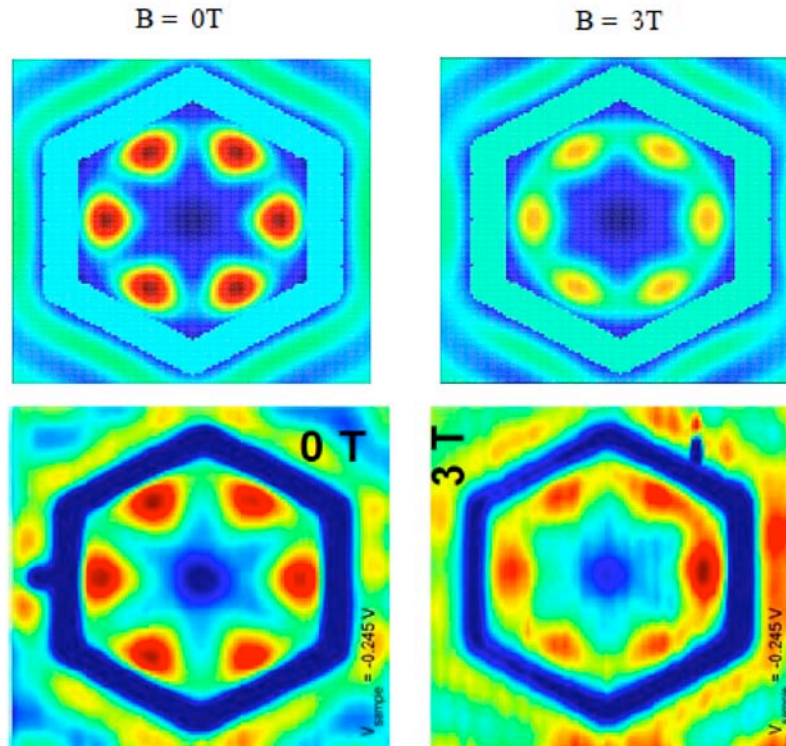
Over a century ago, it was understood that light emitted from atoms comes in certain well defined colors. This led to a scientific revolution when physicists developed quantum theory to explain the spectrum of emitted colors. They discovered that the distinct quantum energy levels of electrons orbiting the nucleus were responsible. These energy levels also exist in solids, and they are especially interesting when electrons are trapped closely together. One way to trap the electrons is using walls made of thin layers of

different materials. This creates “two-dimensional” systems where electrons are unable to move up and down but free to move in a plane, as if they were placed on a sheet of paper. These 2-D electron systems have not only hosted amazing discoveries leading to two Nobel prizes, but they are also used in sensitive high frequency amplifiers. Unfortunately, it has proven difficult to measure their energy levels. A new technique developed in the Ashoori group at MIT allows measurement of their energy spectrum with 1000 times better energy resolution than was previously possible. Researchers use a quantum mechanical effect called tunneling, where electrons are able to pass through barriers that would normally be insurmountable. When electrons hit the walls forming the 2-D system at the correct energy to enter an energy level inside, they are able to tunnel much more quickly than they would otherwise, allowing the spectrum to be mapped by measuring how quickly electrons tunnel at different energies. Without this spectrum, scientists have learned about 2-D systems by disturbing the electrons electrically and looking at what they do. Like putting small ripples on the surface of the sea, this only tells about what is happening at the edge of the states that are filled

in the spectrum. These spectra provide the first glimpse of the entire “sea” in these systems and show what a beautiful and interesting view that can be.

Imaging a Quantum Wave

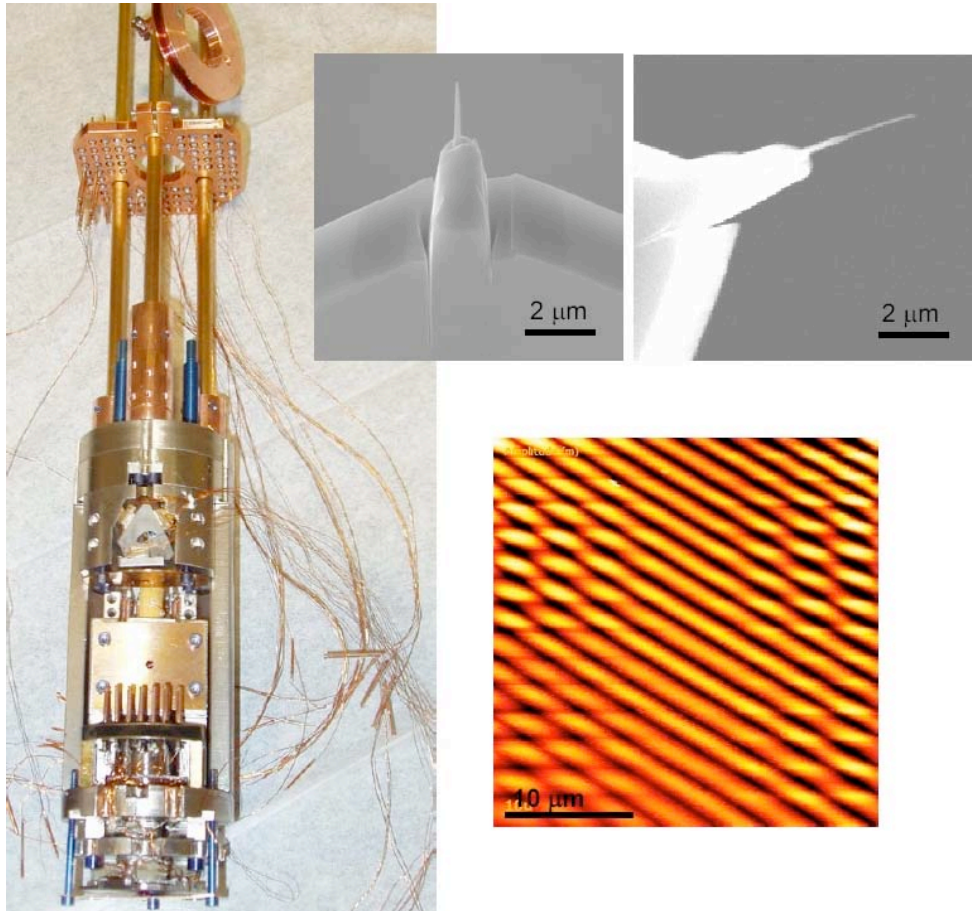
Eric J. Heller and Hari Manoharan



Experimental (bottom) and theoretical (top) nanoscale scanning tunneling microscope images of a hexagonal array of CO molecules (dark blue) forming a box that confines electrons on a (111) surface of copper. On the left, the conductance is shown as a function of the STM tip position, with no magnetic field, and on the right there is a large 3 Tesla magnetic field perpendicular to the image. Good agreement is seen between theory and experiment, which concur on the significant effect of the magnet field. The conductance reflects the pattern of several standing waves of the hexagon, which have nearly the same energy. This represents the direct imaging of a quantum wave, and it advances our knowledge of electron motion and scattering on surfaces, which could become part of lightning fast electronics of the future.

High Resolution Force Microscope

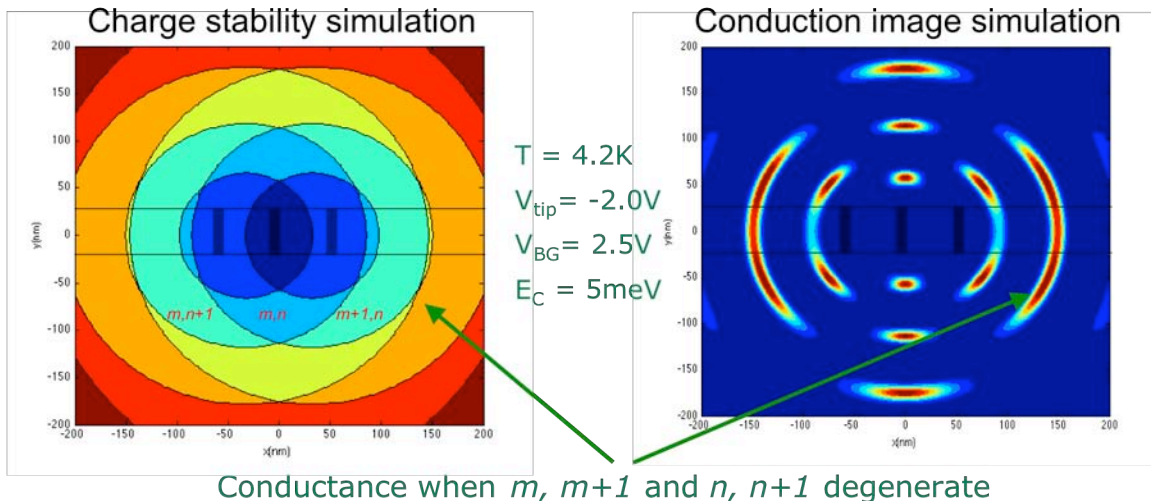
Jennifer Hoffman



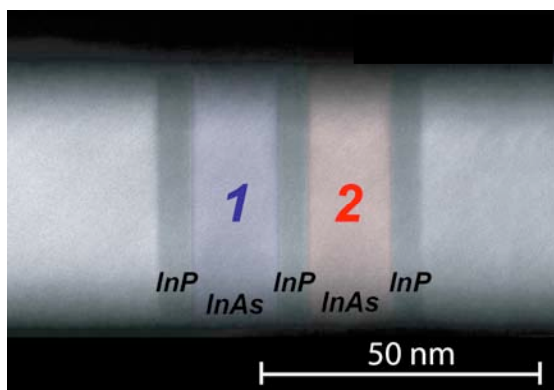
In order to fully exploit the technological potential of superconductors, multi-ferroics, and other novel materials, we need to image and understand magnetic and electric fields at the nanoscale. We have therefore constructed a new cryogenic force microscope with both vertical and horizontal cantilever geometry, to detect and measure magnetic and electrostatic forces parallel and perpendicular to a sample surface, with 20 nanometer spatial resolution and sub-picoNewton force resolution. The collage shows a photo of the assembled microscope, two scanning electron micrographs of cantilever tips (focused ion beam etched silicon tip on the left, and carbon nanotube tip on the right), and the first image acquired with this microscope, of a magnetic hard drive in ambient conditions.

Control of a Double Quantum Dot in an InAs/InP Nanowire by an SPM Tip

R.M. Westervelt and Lars Samuelson



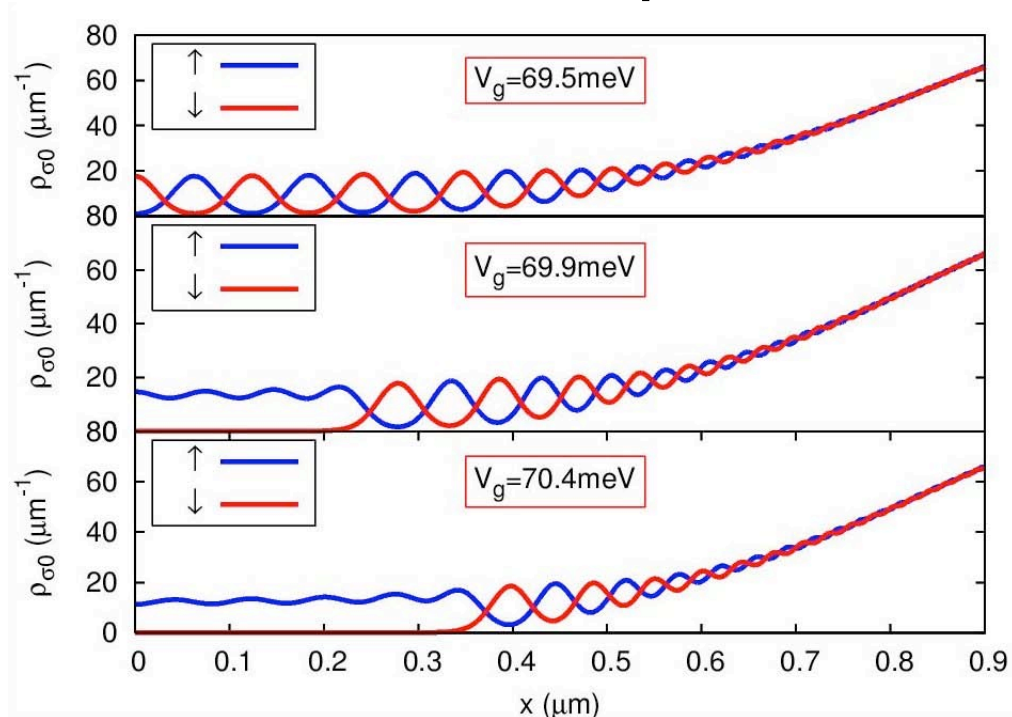
Double quantum dots grown in InAs/InP nanowires are promising for spintronics, due to their tiny size and the electron's large magnetic moment. However their small size makes them difficult to gate individually. The simulations above show that a cooled scanning probe microscope (SPM) tip can be used as a movable gate to control an InAs/InP nanowire double dot. The position of SPM tip can individually tune the charge in each dot, as shown by the overlapping bullseye patterns on the left. Series conduction only occurs at the ring intersections, where the charge on both dots can change, as shown by the simulated SPM conductance image on the right. This image corresponds to the conventional charging diagram for planar double dots obtained by varying the gate voltage of each dot.



TEM image of a tunnel-coupled InAs double dot with InP barriers, grown in an InAs/InP nanowire heterostructure. Optimized growth technique leads to dots of nearly identical size. (Samuelson)

Effects of Electron-Electron Interactions in an Inhomogeneous One-Dimensional Wire

Bertrand I. Halperin



One-dimensional metals, in semiconductor nanowires made from InAs and other materials, are interesting systems for fabrication of nanoscale semiconductor devices, which are under active investigation in experimental and theoretical work supported by our NSEC. Electron-electron interactions can become very important when the electron density is low, and can have subtle effects when this density varies rapidly with position. The Figure shows a Hartree-Fock calculation of the densities of spin-up and spin-down electrons, as a function of position, in a one-dimensional quantum wire, with reduced density near the center due to the influence of a repulsive external gate. Note the transition from antiferromagnetic spin-order to complete spin alignment, in the central region, as the gate potential is increased and the electron density is further reduced. The system is symmetric about $x = 0$.

Figure taken from J. Qian and B.I. Halperin, "Hartree-Fock calculations of finite inhomogeneous quantum wire," arXiv:0707.2992; *Phys. Rev. B* (in press)

Electron and Hole-rich Regions in Graphene

Amir Yacoby

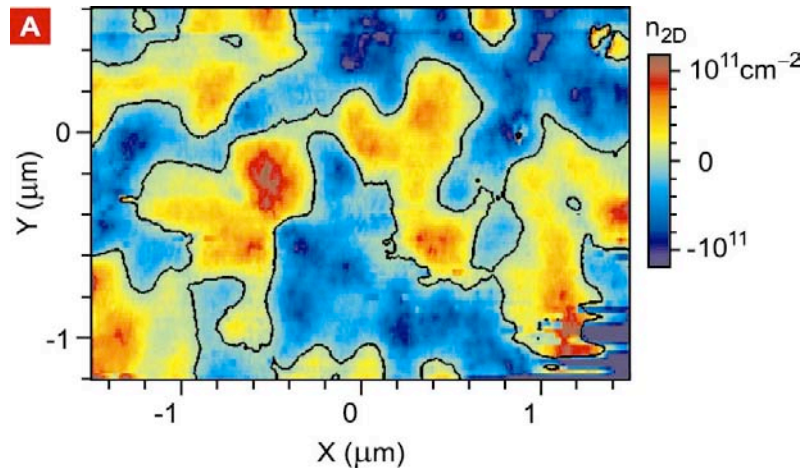


Figure 1. Single electron transistor (SET) image of electron-rich (red) and hole-rich (blue) regions in a graphene layer.

Even under conditions of zero average carrier concentration, the conductivity of graphene is finite. The emergence of puddle-like electron and hole-rich regions in graphene, imaged in the Yacoby group using a scanning single electron transistor, could be the reason for such anomalous minimal conductivity.

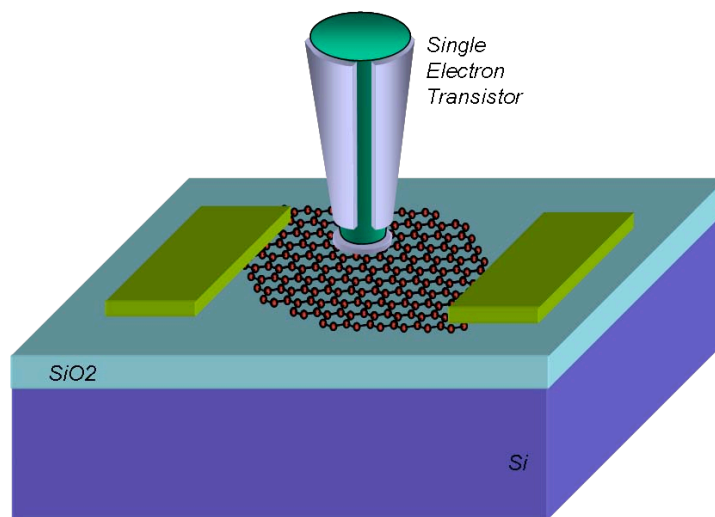
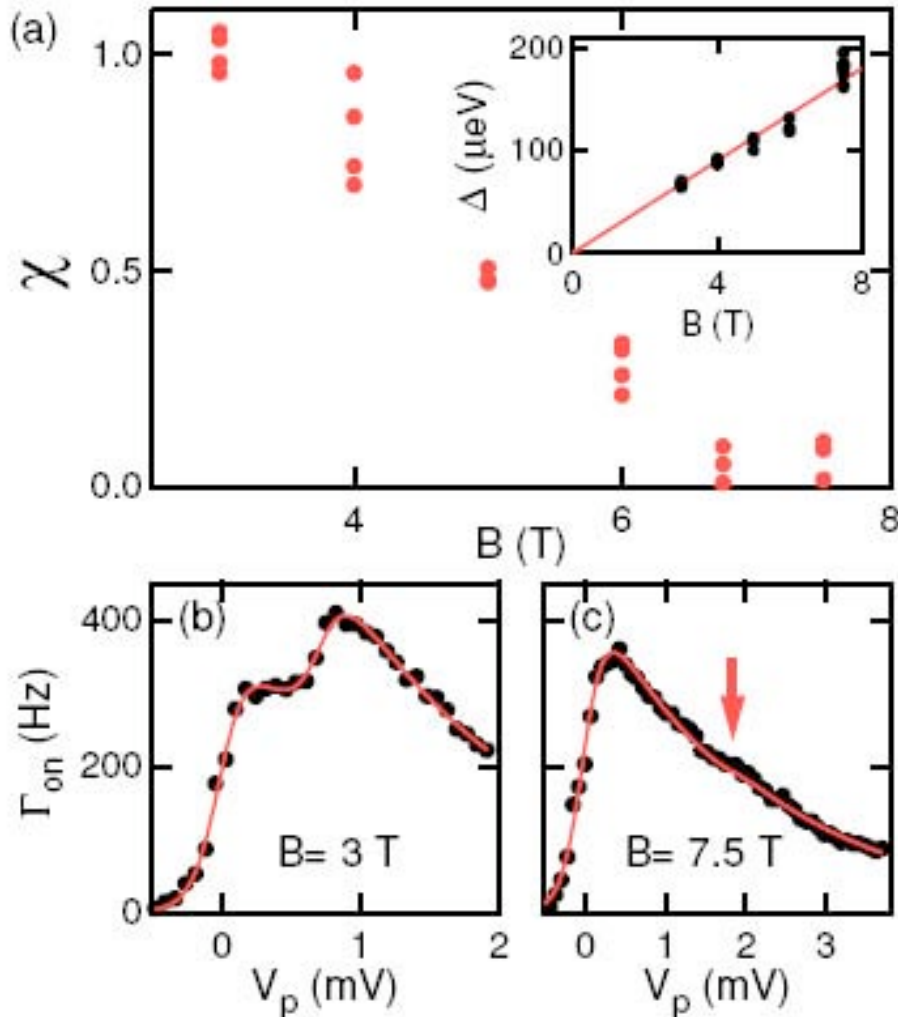


Figure 2. Schematic diagram showing an SET probe above a graphene layer.

Spin-dependent Tunneling

Marc A. Kastner



Devices using the spin of electrons, rather than their charge, have been proposed to reduce power dissipation. For these it is necessary to be able to differentiate the two spin states of electrons. Theory predicts that electrons with the two spin states tunnel at equal rates. However, our experiment shows that this is not always true. At low field (**b**) one sees both spin states, but at high field (**c**) only one. The ratio of the rates for the two states χ is plotted as a function of field in (**a**). While this behavior is not yet understood, it offers the possibility of a new method for filtering the spin states of electrons.

Nanoparticle-Based Molecular Imaging for MRI

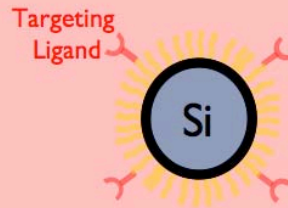
Charles Marcus

Technology Driven ...



Magnetic Resonance Imaging (MRI)

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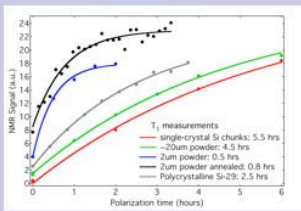
Nanoparticle Imaging Agent (NIA)

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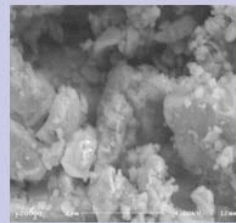
Molecular Contrast

Multidisciplinary ...



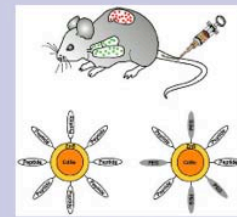
Solid State Physics

- Hyperpolarization to improve the sensitivity of MRI by >10,000x
- Spin polarization lifetimes (T1) of >4h allow longitudinal tracking of nanoparticles *in vivo*



Nanoscale Fabrication and Material Science

- Efficient, scalable production of high-resistivity nanoparticles
- Ex vivo* analysis of gross and nuclear magnetic properties



Biological Targeting

- Functionalization for selective uptake by certain tumors.
- Analysis of nanoparticle kinetics and toxicity *in vivo*.

Sweet Spot in Double Semiconductor Quantum Dot Electronic Structure

Michael Stopa

The field of quantum computation is in its infancy and numerous physical systems have been proposed as platforms for implementing quantum algorithms. A necessary criterium for such computation is the preservation of coherence at the quantum level. This coherence requires considerable control and, typically, very low temperatures.

In the interaction between qubits in semiconductor quantum dot systems, the mutual rotation of two spins, in adjacent dots, is controlled by the exchange interaction. Unfortunately, the strength of this exchange coupling is extremely sensitive to small variations in the coupling between the two dots and therefore to small fluctuations in the electrostatic environment.

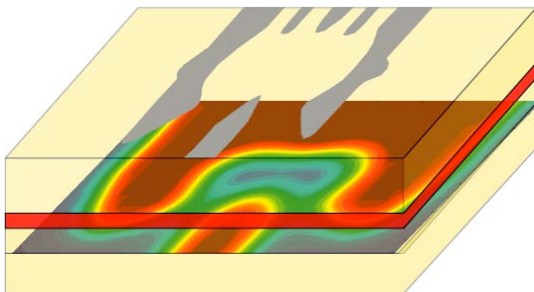


Figure 2. The gate pattern and wafer profile of the double dot are illustrated here, along with the self-consistent potential profile in the 2-D electron gas.

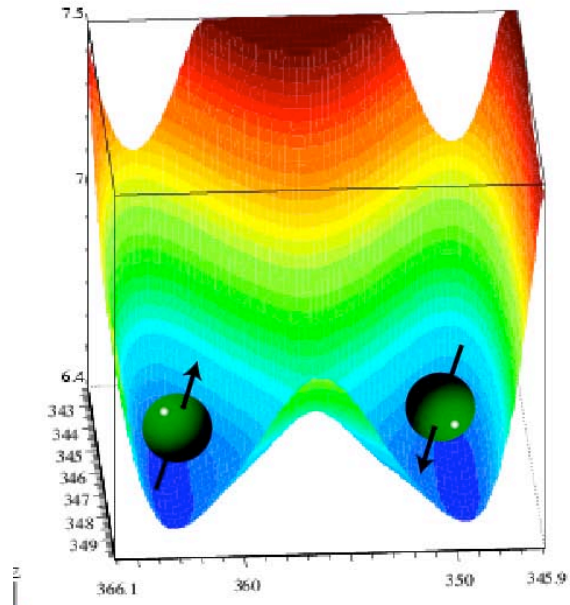


Figure 1. Two electrons, confined in adjacent semiconductor quantum dots, interact through the exchange interaction and thereby mutually rotate their spins in a fundamental process for quantum computation.

We have recently used our configuration interaction simulations to model “artificial molecular hydrogen” (the fundamental two-qubit system) in a semiconductor double dot. We have been able to locate a region in parameter space, and elucidate the reason for the existence of such a region, where to lowest order the sensitivity of the system to electronic (voltage and charge) fluctuations vanishes.