

Editors: Sokrates T. Pantelides, Mark A. Reed, James S. Murday and Ari Aviram Frontmatter

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# **Molecular Electronics**



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# MATERIALS RESEARCH SOCIETY SYMPOSIUM PROCEEDINGS VOLUME 582

# **Molecular Electronics**

Symposium held November 29-December 2, 1999, Boston, Massachusetts, U.S.A.

## **EDITORS:**

# Sokrates T. Pantelides

Vanderbilt University Nashville, Tennessee, U.S.A.

# Mark A. Reed

Yale University
New Haven, Connecticut, U.S.A.

# James S. Murday

Naval Research Laboratory Washington, D.C., U.S.A.

### Ari Aviram

IBM T.J. Watson Research Center Yorktown Heights, New York, U.S.A.



Materials Research Society

Warrendale, Pennsylvania



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### **PREFACE**

This volume contains papers presented at Symposium H, "Molecular Electronics," held November 29-December 2 at the 1999 MRS Fall Meeting in Boston, Massachusetts.

As Si-based microelectronics marches to ever-smaller feature sizes, the point is rapidly approaching when conventional MOSFETs will no longer operate. Such devices depend on the concept of "doped regions" which can be envisioned to be ever so small until you get to the point where there is only a handful of dopant atoms. Tunneling across ultra-thin films is another major issue.

Single molecules as active device elements is a concept that goes back to the 1970's, but in recent years very substantial progress has been made in synthesizing molecules suitable for such purposes. Carbon nanotubes have been a prime candidate for both nanowires and active devices. Molecules as small as single benzene rings, and larger, more complex molecules have been synthesized and measured, showing reproducible current-voltage characteristics. Unusual features are large negative differential resistance and large shifts of resonant peaks as functions of temperature.

The symposium featured many invited talks and a fair number of contributed talks; unfortunately many of the invited speakers did not contribute a paper to the proceedings. The interested reader may refer to relevant recent journals where original publications can be found.

The Symposium was extremely well attended by overflow crowds to the point that a closed-circuit TV had to be setup in an adjacent room. It was a very diverse audience, mainstream Si engineers, chemists, physicists, materials scientists—it was a truly interdisciplinary celebration of an exciting emerging field.

We gratefully acknowledge support from the Office of Naval Research.

Sokrates T. Pantelides Mark A. Reed James S. Murday Ari Aviram

September 2000



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SESSION H1: Chair: Sokrates T. Pantelides Monday Morning, November 29, 1999 Cape Cod/Hyannis (M)

8:30 AM \*H.1 OPPORTUNITIES FOR MOLECULAR ELECTRONICS IN HIGH-PERFORMANCE COMPUTER SYSTEMS. <u>Rick Lytel,</u> Howard Davidson, Nyles Nettleton, Sun Microsystems, Palo Alto, CA.

Silicon-based VLSI continues to scale to smaller dimensions and higher performance. This scaling should continue for about ten more years. At that point, new approaches for designing and fabricating CPU and memory devices will be required. This opens the door for the introduction of new technologies based upon molecular chemistry and solid-state physics. But such opportunities exist now in select areas, such as large, fast cache memories. This talk will review the scaling behavior of modern computer systems, will describe opportunities for molecular electronics within the next decade of Silicon scaling, and will examine possible designs for future computing machines based upon molecular electronics.

9:00 AM \*H1.2 WIRES, SWITCHES, AND WIRING: TOWARD A CHEMICALLY ASSEMBLED ELECTRONIC NANOCOMPUTER. J. Heath, Dept. of Chemistry and Biochemistry, University of California at Los Angeles, Los Angeles, Los Angeles, Los Angeles, Los

Researchers at UCLA, Hewlett Packard, and UC Berkeley have begun to fabricate a molecular based computational machine. Architectural schemes and rationale for this machine will be presented. The fabrication and characterization of electronically configurable logic gates and logic circuits based on molecular-scale switches and nano-scale wires will be presented. Fundamental issues related to these devices will be discussed, including interface issues, switching times, and molecular structure optimization.

9:30 AM \*H1.3 NANOTUBE DOTS, WIRES, AND CROSSES. Paul L. McEuen, Dept. of Physics, Univ. of Calif. -Berkeley, and Materials Sciences Division, LBNL.

Carbon nanotubes are a new class of one-dimensional conductors that can be either metallic or semiconducting, depending upon their structural details. In this talk, I will discuss experiments to probe the electrical properties of these fascinating systems. Whese are attached to individual tubes and a nearby gate is used to control the charge per unit length of the tube. If the tube is semiconducting, the device operates as the world's smallest transistor. For a metallic tube, a correlated electron state known as a Luttinger liquid is found. Short tubes act like one-dimensional quantum dots whose discrete energy levels and spin states can be probed. Crossed metal and semiconducting tubes act as ultra-small Schottky diodes. As these experiments show, nanotubes offer an unprecedented opportunity to explore the physics and technology of 1D conductors. Carbon nanotubes are a new class of one-dimensional conductors that

SESSION H2: Chair: Sandra J. Rosenthal Monday Morning, November 29, Cape Cod/Hyannis (M)

10:30 AM \*H2.1 NANOCRYSTALS AS BUILDING BLOCKS FOR ELECTRICAL DEVICES. Paul Alivinatos, Univ of California, Berkeley, Dept of Chemistry, Berkeley, CA.

Nanometer size crystals of inorganic semiconductors display a wealth of size dependent optical and electrical characteristics. Such nanocrystals (CdS, CdSe, InP, InAs), with well controlled interfaces, can now be fabricated by colloidal chemistry techniques. The resulting particles are extremely flexible chemically, and may be thought of as class of molecule. These nanocrystals can in turn be incorporated into electrical devices. Three examples will be described: EDBa, photovoltaics, and single nanocrystal transistors. Finally, efforts will be described to use biological molecules, such as DNA, to design complex patterns of nanocrystals.

11:00 AM H2.2 ELECTRONIC TRANSPORT IN COLLOIDAL CDSE QUANTUM DOT ARRAYS. N.Y. Morgan, I. Prasad, M.A. Kastner, M.I.T., Dept. of Physics, Cambridge, MA, C.A. Leatherdale, M.G. Bawendi, M.I.T., Dept. of Chemistry, Cambridge, MA.

We present results for dark current measurements on large close-packed arrays of CdSe nanocrystals. The nanocrystals are

chemically synthesized in solution and then deposited between gold electrodes onto a silicon substrate which has a 350 nm oxide grown on its surface. The Si substrate is degenerately doped and can be used as gate. In response to an applied voltage step, we observe a power-law decay of the current over five orders of magnitude in time and four orders of magnitude in current. Furthermore, we do not observe a steady-state dark current for fields up to 1 x 10 $^9$  V/cm and times out to 5 x 10 $^5$  seconds. We see clear evidence for the injection of electrons into the array, but do not observe hole injection. The number of electrons in the array can be increased by cooling the sample with a positive gate voltage. However, as the number of electrons is increased, the magnitude of the current transient decreases substantially. We believe our results can be understood in the context of the Coulomb glass model, where transport is dominated by the long-range electrostatic interactions between electrons at different sites.

11:15 AM H2.3

PRESSURE-INDUCED INTERDOT INTERACTIONS IN Cdse
NANOCRYSTAL ARRAYS. Bosang Kim, Mohammad A. Islam, Louis

E. Brus and Irving P. Herman, Columbia Radiation Laboratory,
Columbia University, New York, NY.

Columbia University, New York, NY.

Three-dimensional arrays of organically-passivated CdSe nanocrystals were investigated under hydrostatic pressure using photoluminescence and absorption spectroscopy. In some arrays, such as those composed of CdSe dots capped by tien-noctylphosphine oxide, the first excitonic level of the absorption spectrum monotonically moves to the blue with increasing pressure until the structural phase transition is reached, reflecting the increasing band gap of bulk CdSe and the increasing confinement energy. This dependence was also seen in dilute solutions of CdSe nanocrystals. For arrays composed of CdSe dots capped with some smaller molecules, the energy of the first excitonic peak in absorption does not increase with pressure above ~40 kbar. This may be evidence of interdot coupling. This work was supported by the MRSEC Program of the National Science Foundation under Award Number DMR-9809867 and the JSEP Program under Contract No. DAA-GSS-97-1-0166.

11:30 AM H2.4

ORGANOMETALLIC SYNTHESIS AND SPECTROSCOPIC

CHARACTERIZATION OF MN DOPED CDSE NANOCRYSTALS.

Frederic V. Mikulee, Masaru Kuno, Moungi G. Bawendi, MIT, Dept.

of Chemistry, Cambridge, MA: Marina Bennati, Dennis A. Hall,

Robert G. Griffin, MIT, Dept. of Chemistry and Center for Magnetic

Resonance, Francis Bitter Magnet Laboratory, Cambridge, MA.

Resonance, Francis Bitter Magnet Laboratory, Cambridge, MA.

The synthesis of II-VI semiconductor nanocrystals doped with transition metals has proved to be particularly difficult. In the case of CdSe quantum dots produced via high temperature pyrolysis in trioctylphosphine oxide (TOPO), specially designed precursors used in this study appear to be necessary to successfully incorporate low levels of Mn. Combining a simple etching experiment and electron paramagnetic resonance (EPR) measurements reveals that most of the dopant atoms reside inside the surface layers of the inorganic lattice. The dopant dramatically affects Cd-13 magic angle spinning (MAS) NMR spectra; the observed paramagnetic shift and decreased longitudinal relaxation time reproduce bulk material behavior. Results from fluorescence line marrowing (FLN) studies on Mn doped CdSe nanocrystals mirror previous findings on undoped nanocrystals in an external magnetic field. Experimental fitting of photoluminescence excitation (PLE) spectra of doped nanocrystals reveals that the effective absorption lineshape contains a new feature which is believed to be a previously unobserved - but theoretically predicted - optically dark fine structure state.

11:45 AM <u>H2.B</u>
ARTIFICIAL ATOMS OF SILICON. Justin D. Holmes, Kirk Ziegler,
Keith P. Johnston, Chris Doty, <u>Brian A. Korgel</u>, Dept of Chemical
Engineering and the Texas Materials Institute, The University of
Texas, Austin, TX.

Highly size monodisperse, sterically-stabilized silicon nanocrystals ~13 Å in diameter were synthesized using a new solution-phase synthetic approach that utilizes a capping solvent heated and pressurized above its critical point to control particle growth. The precursor diphenylsilane was thermally degraded in octanol at temperatures (773 K) and pressures (340 atm) well above the critical point for octanol: 34 atm and 656 K. These nanocrystals represent the first examples of silicon artificial atoms that exhibit discrete optical transitions in the room temperature absorbance and photoluminescence excitation spectra with efficient band edge photoemission (quantum yield=23%). Infra-red and chemical ionization mass spectroscopies indicate that the stabilizing C8 hydrocarbon chains bind to surface-exposed silicon atoms through an Si-O-C link at the particle surface. These silicon clusters, on the lower size limit, reveal that silicon does not undergo the transition to a direct band gap semiconductor and furthermore, that excitons in

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these very small clusters remain strongly coupled to phonons, resulting in broad luminescence at room temperature.

SESSION H3: Chair: Norton D. Lang Monday Afternoon, November 29, 1999 Cape Cod/Hyannis (M)

1:30 PM \*H3.1
NANOSTRUCTURED SELF-ASSEMBLED MONOLAYERS FOR MOLECULAR ELECTRONICS. Chongwu Zhou, M.R. Deshpande and M.A. Reed, Dept of Electrical Engineering, Yale University, New Haven, CT; L. Jones III, J. Seminario and J.M. Tour, Dept of Chemistry, Univ of South Carolina, Columbia, SC.

The measurements of electronic transport in organic molecular wires are experimentally challenging and intriguing. We present the investigation of novel metal / self-assembled monolayer of molecular wires / metal heterostructures with nanoscale device areas. Diode-like I-V characteristics is observed for devices consisting of 4-thio-actylibiphenyl and variable temperature measurements reveals the transport mechanisms. A sharp decrease in conductivity is observed around 25 K for devices consisting of molecules with triples bonds in the conjugated backbone. Theoretical simulations suggest it can be explained through thermally excited bond rotations at the triple bonds. Results with other molecules will also be presented.

2:00 PM H3.2
NEGATIVE DIFFERENTIAL RESISTANCE IN NANOSCALE
MOLECULAR JUNCTIONS. Jis Chen, Mark A. Reed, Yale
University, Dept of Electrical Engineering, New Haven, CT; Adam M.
Rawlett, James M. Tour, Rice University, Center for Nanoscale
Science and Technology, Houston, TX.

We report the first observations of negative differential resistance (NDR) with giant peak-to-valley ratio (PVR) in nanoscale molecular junctions. We have measured a record 1000:1 PVR in a 1-thioacetyo-4 (4-(phenylethylny)-2-amino-5-nitro-1/cethynyl) phenyl) benzene self-assembled monolayer (SAM) between metal contacts. Variable temperature measurements show that the junction develops NDR below 220 K, reaching a maximum negative differential conductivity of -5 KS /cm² at 2 V, and an associated PVR of 1000:1 at 60 K. The role of the internal redox dipole of the molecule on the conduction, and methods to engineer the operation for 300 K, will be discussed. The device exhibits the largest PVR known for any structure with useful (>>KT) operating voltages.

2:15 PM. \*H3.3
TRANSPORT CALCULATIONS IN MOLECULAR DEVICES
FROM FIRST-PRINCIPLES. M. DI Ventra, N.D. Lang, and S.T.
Pantelides! 'Department of Physics and Astronomy, Vanderbit
University, Nashville, TN; 'IBM Research Division, Thomas J.
Watson Research Center, Yorktown Heights, NY.

Watson Research Center, Yorktown Heights, NY.

For molecular electronics, Boltzmann's equation is no longer valid for simulating device characteristics. We present the first fully ab-initio simulation of a molecular device that has already been studied experimentally by Reed et al. [1], namely a benzene-1,4-dithiolare molecule between gold electrodes. The theoretical I-V curve has the same overall shape as the experimental curve – reflecting the electronic structure of the molecule in the presence of the electric field – but the absolute value of the current is very sensitive to contact chemistry and geometry. In particular the presence of a single gold atom between the molecule and the electrode surface reduces the conductance by about two orders of magnitude. Replacement of the single gold atom by an aluminum atom, whose p orbitals couple more effectively to the molecule's p orbitals, increases the conductance by about an order of magnitude. We also show the polarization induced by a third terminal (gate) on the above device. Current gain due to the gate bias can be achieved at reasonable gate fields. Finally, the effect of current-induced forces on the device will be discussed.

[1] M.A. Reed et al., Science 278, 252 (1997)

Work supported in part by DARPA.

2:45 PM H3.4

ELECTRICAL TRANSPORT MEASUREMENTS THROUGH A

METAL-MODLECULE(S)-METAL JUNCTION. J.P. Bourgoin¹, C.

Kergueris¹, S. Palacin¹, D. Esteve², C. Urbina², M. Magoga², C.

Joachim³, ¹Service de Chimie Moléculaire, CEA Saclay, FRANCE;

²Service de Phyaque de l'Etat Condensé, CEA Saclay, FRANCE;

²CEMES/LOE CNRS Toulouse, FRANCE.

In order to measure the transport properties of molecules we have used microfabricated break junctions as metallic tweezers with a nanometer scale adjustable gap.[1] Specifically designed molecules,

bisthiololigothiophene in particular, were self-assembled in the gap to form metal-molecule(s)-metal junctions. The current-voltage characteristics recorded at room temperature are non-linear with step-like features and can be strongly modified by gap size changes of 0.4 Å/2] These results can be qualitatively accounted for by a model which assumes that a single molecule is involved and that takes explicitly into account its discrete electronic levels. Finally, we will compare theoretically thiol and isocyanide binding groups in terms of their electrical transport efficiency. [1] M.A. Reed, C. Zhou, C.J. Muller, T.P. Burgin, J.M. Tour, Science 5336 (1997) 232 [2] C. Kergueris, J.P. Bourgoin, S. Palacin, D. Esteve, C. Urbina, M. Magoga, C. Joachim, Phys. Rev. B 59 (1999) 12505

SESSION H4: Chair: Mark A. Reed Monday Afternoon, November 29, 1999 Cape Cod/Hyannis (M)

3:30 PM H4.1

NANOMETER SIZED GAPS IN METALLIC WIRES.

M. Radosavijevic, J. Lefebvre, A.T. Johnson, Dept. of Physics,
University of Pennsylvania, Philadelphia, PA.

We report on the details of a new method for production of nanometer sized breaks (nanogaps) in metallic wires. Our method, based on standard lithographic techniques, offers possibility of parallel production in an industrial setting. In addition, we can accurately tune the size of individual nanogaps: from 100nm to below 5nm. Such small separations exceed the limits of conventional electron beam lithography, and compare favorably to other, more involved, schemes. Preliminary spectroscopic data on SWNTs bridging a nanogap are presented and analyzed in the single electron charging picture.

3:45 PM H4.2

NANOELECTRODE FABRICATION AND SELF-ASSEMBLY OF
SINGLE ELECTRON TRANSISTOR. Magnus Persson, Linda
Olofsson, Chalmers University of Technology, Dept. of
Microelectronics and Nanoscience, Gothenburg, SWEDEN; Charles M.
Marcus, Alberto Morpurgo, Stanford University, Dept. of Physics, CA.

marcus, aloerto Morpurgo, Stanford University, Dept. of Physics, CA. Angle evaporation and controlled electrodeposition of electrodes have been used to fabricate very small well-defined gaps between two gold electrodes. The size of the gap can be controlled between 1-10 nm or less, which is much smaller than one could achieve by only using angle evaporation and lift-off technique. The conductance is measured during the process and conductance quantization is observed, indicating nanobridges with only single or few gold atoms. The electrodes have been used for connecting ligand stabilized gold clusters, with 2-5 nm diameter, which are self-assembled in the electrode gap. Coulomb blockade was observed at 7-2 K, in such devices. The well-controlled gap can also be used for contacting self-assembling conducting oligomers. The electrodes are made on thin Silicon-nitride membranes and are studied with a transmission electron microscope.

4:00 PM \*H4.3 STM PROBING OF QUANTUM AND MOLECULAR MECHANICS OF INDIVIDUAL MOLECULES. <u>James K. Gimzewski</u>, IBM Research Division, Zurich Research Laboratory, SWITZERLAND.

Research Division, Zurich Research Laboratory, SWITZERLAND.

On the scale of individual molecules adsorbed on surfaces both mechanics and quantum effects combined with thermal fluctuations can play a determining role in their electronic transport proerties and their dynamics. These effects, rather than limit future electronic devices, may provide unique oppoprunities to design and build new machines. The nanoscale science underlying electronic and mechanical properties of single meolecules is at an early stage of developement. Nevertheless the capabilities of designer chemical synthesis opens mey opportunites to design experiments within a single molecule. In this talk we will discuss two molecules that were designed to: (1) Understand the role of a molecule in extending a metallic wavefunction and therby providing electrical conduction at low voltages and (2) Demonstrate the rotation of a single molecule operating within a surpramolecular bearing. We extend both of these concepts with a new proposal for a type of electronic device that utilises quantum electronic transport modulated by Brownian rotaion fluctuations. The redirection of the fluctuations is achieved by the application of either coloured noise or by an external static pertubation.

4:30 PM \*H4.4 THE CONDUCTANCE OF A MOLECULAR WIRE. Christian Joachim, University of Toulouse, CEMES-CNRS, Toulouse, FRANCE.



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The conductance of a molecular wire is a decreasing exponential function of its length. This defines the tunnel transport regime where the homo-lumo gap of the molecular wire is different from zero and the I-V characteristic is linear at low voltage. We will first show how to optimize the chemical structure of the molecular wire to reach a tunneling inverse damping length hower than 0.1 Ang-1. Then, we will report an STM experiment to mesure this damping length on molecular wires 0.3 mm in width, 1.4 and 4 mm in length in a planar configuration using the STM tip like a stub scanned along the molecular wire. The properties of intramolecular circuits will be discussed in conclusion.

SESSION H5: Chair: Bruce E. Gnade Tuesday Morning, November 30 Cape Cod/Hyannis (M) 30, 1999

8:30 AM \*H5.1 NOVEL STRATEGIES FOR PREPARATION OF ORDERED ARRAYS OF ORGANIC MOLECULES ON SEMICONDUCTOR SURFACES. Robert J. Hamers, University of Wisconsin, Dept. of Chemistry, Madison, WI.

SURFACES. Robert J. Hamers. University of Wisconsin, Dept. of Chemistry, Madison, WI.

We have developed a new strategy for forming ordered, oriented organic layers on (001) surfaces of group IV semiconductors. Our strategy is based on the transfer of directional bonding from a solid inorganic substrate into an attached array of organic molecules. Si(001), Ge(001), and C(001) (diamond) surfaces consist of Siz-Si, Ge=Ge, or C=C dimers, with the atoms within each dimer held tagether by a strong sigma and weak pi bond. Unsaturated organic compounds (containing one or more double or triple bonds) occur rapidly on these surfaces, producing well-defined, stomically-sharp organic-inorganic interfaces. One important property of organic layers formed using this chemistry is that the molecules are oriented. This orientation arises because the bonding of the molecule to the surface involves two bonds; thus, the orientation of the Siz-Si dimers of the underlying substrate is transferred into the orientation of the organic monolayer films. The orientation of the surface dimers can be controlled by using vicinal surfaces. For example, on silicon (001) miscut by 4 degrees toward < 110 >, a single domain surface is formed in which all the dimers are aligned along the same direction; the molecules in the organic film are also therefore aligned. The alignment of organic molecules provides anisotropy in their physical properties such as optical absorption and, in principle, electrical onductivity. STM measurements of the apparent height of individual molecules also provides insight into the mechanisms of electrical onductivity and the effects of various substituent groups on transport from the surface dimers of electrical onductivity and the effects of various substituent groups on transport functional groups; this permits the formation of novel structures that may have unusual chemical, optical and/or electrical properties by virtue of their spatial orientation. This talk will discuss our recent efforts at fabrication of highly-

9:00 AM H5.2
STM:INDUCED PHOTON EMISSION FROM SINGLE
PORPHYRIN MOLECULES ON Cu(100), Z.-C. Dong, T. Ohgi, D.
Fujita, T. Yakabe, H. Nejoh, T. Terui, S. Yokoyama, M.-N. Zhou, S.
Mashiko, T. Okamoto, Natl Research Inst for Metals, Ultrahigh
Vacuum Station, Tsukuba, JAPAN.

Positioning of a scanning tunneling microscopy tip above a single Cu-TBP porphyrin molecule on Cu(100) is found to induce photon emission when electrons are injected and bias voltages are above ±2 V. The emitted light appears orange by the naked eyes, in agreement with a broad spectral peak around 650 mm. Amazingly, this broad peak features several shoulder-peaks with a peak spacing of ca. 800 cm<sup>-1</sup>, an implication of the fine structure related to the vibrational excitation of the molecule. The intensity of light emission is measured to be about 10<sup>-8</sup> cps, resulting in a quantum efficiency in the order of 10<sup>-8</sup>. A new approach for producing strong light emission with control will be proposed.

9:15 AM H5.3
FABRICATION AND CHARACTERIZATION OF
MONOMOLECULAR SIGMA/PI HETEROSTRUCTURES. J.
Collet, S. Lenfant, <u>D. Vuillaume</u>, Institut d'Electronique et de
Micro-Électronique du Nord, CNRS, Villeneuve d'Ascq, France. O.
Boulousa, F. Rondelez, Physicochimie Curie, Institut Curie, CNRS,
Paris, FRANCE; J.M. Gay, Centre de Recherche sur les Mécanismes
de la Croissance Crystalline, CNRS, Marseille, FRANCE; K. Kham,
C. Chevrot, Laboratoire de Recherche sur les Polymères et Matèriaux

Electro-actifs, Univ. Cergy-Pontoise, FRANCE.

Electro-actifs, Univ. Cergy-Pontoise, FRANCE.

Molecular-scale Electronics is envisioned as a promising bottom-up strategy to nanoscopic electronic devices. The use of self-assembly etchnique is an invaluable strategy to fabricate such structures. Self-assembled monolayers (SAM) of saturated alkyl chains for instance are known to present a very good insulating behavior. Here, we have used a chemical functionalization of the end-groups of such organic SAM's to fabricate insulator/semiconductor heterostructures (hereafter called sigma-pi SAM) at a molecular level. By attaching pi-electron moleties at the top of previously formed insulating monolayers, we obtain a reasonably large in-plane conductivity without hampering the insulating behavior in the perpendicular circution. The ratio between the parallel and perpendicular conductivities is found to be very large, up to 128 for +p monolayers terminated by various pi moseties like Phenyl, Retinol, Pyrene, thiophene, anthracene and Carbazole. Two chemical routes are described to build these structures. FTIR, wettability, ellipsometry and X-ray reflectivity carefully characterized the structural properties of these SAMs before electrical measurements. The in-plane conductivities in these SAMs were analyzed versus the electronic structures of the pi end-groups. Nature of the conduction mechanism will be discussed. In many proposals for molecular-scale devices, the sigma-pi interface is proposed as a way to modulate the electron transfer (blocking barrier). This concept is demonstrated here at the monolayer level. Such insulator/semiconductor heterostructures with a relatively high anisotropic conductivity open powerful opportunities to build future nanoscopic molecular devices.

9:30 AM HS.4
ELECTRONIC STRUCTURE OF THIENYLENEVINYLENE
OLIGOMERS: APPLICATION TO THEIR ADSORPTION ON
SILICON (100) SURFACE. C. Krzeminski, C. Delerue, G. Allan, V
Haguet, B. Grandidier, J.P. Nya, D. Stievenard, IEMN, Depl ISEN.
Lille, FRANCE; P. Frère, E. Levillain, J. Roncali, Ingénieire
Moléculaire et Matériaux Organiques, Université d'Angers, Angers,
PERANCE; P. Prère, E. Levillain, J. Roncali, Ingénieire

FRANCE.

Conjugated oligomers have attracted considerable interest as possible molecular wires. Investigation of the interaction of these molecules with the silicon (100) surface is therefore useful to gain insight into the way the oligomers could be connected to the silicon surface. We present combined experimental and theoretical studies on such a system. Theoretically, ab initio calculations based on the local density approximation and semi-empirical tight binding calculations are applied to thienylenevinylene oligomers up to the hexadecamer stage (m=16). The results correctly describe the experimental variations of the gap versus size, the optical spectra and the electrochemical redox potentials. We propose a simple model to deduce from the band structure of the polymer chain the electronic states of the oligomers close to the gap. We analyze the evolution of the gap as function of the torsion angle between consecutive cells: the modifications are found to be small up to a ~30° angle. We show that these oligomers possess extensive pi-electron delocalization along the molecular backbone which supports their interest for future electronic applications such as molecular wires. Experimentally, the adsorption of different thenylenevinylenes on silicon (100) 2x1 have been studied by scanning tunneling microscopy in ultra high vacuum. The current constant images of the thienylenevinylenes are strongly bias dependent and allow the determination of the adsorption sites. The reaction involves the thiophene ring with a single Si atom of the Si=Si dimer. Ab initio calculations are performed to better characterize the chemical nature of the bonding.

9:45 AM H5.5

SELF-ASSEMBLY GROWTH OF MOLECULAR WIRES BY MOLECULAR BEAM DEPOSITION. Chengthi Cai, Johannes Barth, Jens Weckesser, Martin Boesch, Christian Bosshard, L. Buergi, O. Jeandupeux, Peter Guenter, Klaus Kern, ETH, Inst of Quantum Electronics, Zurich, SWITZERLAND; EPFL, Inst of Experimental Physics, Lausanne, SWITZERLAND.

The combination of properly designed supramolecular assemblies and molecular beam deposition is an efficient way to fabricate nanostructures such as molecular wires for potential applications in molecular electronics. We have designed a series of supramolecular polymers in which the monomeric units are linked in a linear and head-to-tail fashion via strong hydrogen bonding. One example is 4-trans-2-(pyrid-4-yl-vinyl) benzoic acid (PVBA). The strong had-to-tail hydrogen bonding was indicated by solid state 15N-NMR studies. Thin films of PVBA can be easily grown on glass substrates by organic molecular beam deposition (OMBD) with an oblique incidence. As shown by second harmonic generation experiments, the films have a directional order that is parallel to the projection of the molecular beam direction on the substrate surface. Moreover, the

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directional order is independent of film thickness at least up to 400 nm, and thermally stable up to 190% C. OMBD of PVBA molecules on Ag(111) surface was studied by low temperature STM. The results reveal that PVBA molecules self-assemble into 1-dimensional wires of 1 nm width along < 112 > directions of the Ag-lattice with mesoscopic ordering at the micrometer scale. The individual 'molecular wires' consist of twinned PVBA rows stabilized by H-bonding. Distance between the parallel wires is at least up to 10 nm. Therefore, we have shown a new way to fabricate micrometer-long molecular wires.

SESSION H6: Chair: Supriyo Datta Tuesday Morning, November 30, 1999 Cape Cod/Hyannis (M)

10:30 AM <u>H6.1</u>
SCANNING POTENTIOMETRY STUDIES OF CHARGE
TRANSPORT IN ORGANIC SEMICONDUCTOR BASED FETS
USING CONDUCTING PROBE ATOMIC FORCE MICROSCOPY
(CP-AFM). <u>Kannan Seshadri</u>, C. Daniel Friable, Department of
Chemical Engineering and Materials Science, University of Minnesota,
Minneapolis, MN.

Minneapoiis, MN.

Investigation of charge transport mechanisms in organic semiconductors is of immense importance to development of devices such as thin film transistors and organic-based LEDs. Of particular interest are as-extitiophene (8T) and pentacene, with hole mobilities high enough for flexible, all plastic devices. Conducting Probe Atomic Force Microscopy (CP-AFM) has been used to measure electrical transport characteristics of 6T. The semiconductor was deposited as crystals, ranging from 1:0 8 molecules (2-14 nm) in thickness and from 1:2 µm in diameter by vacuum sublimation onto SiO<sub>2</sub>/p-Si substrates, with lithographically defined Au contacts. The semiconductor crystallite grows across the gap between two electrodes, with a capacitively coupled gate electrode, oa as to achieve a field-effect transistor configuration (FET). An Au-coated conducting AFM probe is brought into contact with the electrically biased 6T crystallite, and the potential is measured as a function of position. This is a scanning potentiometry experiment, involving mapping the local potential over the surface of the organic semiconductor. Pentacene based FETs were also studied by fabricating contacts atop entacene films on Si substrates, representing a top-down approach in FET configuration. Mesoscopic transport measurements, in combination with AFM maging, are a useful strategy for elucidating charge transport across grain boundaries, charge trapping, and other structure-transport relationships in organic materials.

10:45 AM H6.2 CONTACT RESISTANCE MEASUREMENTS ON INDIVIDUAL GRAINS OF a SEXITHIOPHENE. Anna Chwang, C. Daniel Frisbie, University of Minnesota, Dept of Chemical Engineering and Materials Science, Minneapolis, MN.

We describe four point probe measurements on individual grains of the molecular semiconductor sexithiophene (6T). These measurements utilize thin 6T grains (2-14 nm in thickness and 1-2  $\mu$ m in length and width) deposited by vacuum sublimation onto SiO<sub>2</sub>/Si substrates previously patterned with sets of four closely spaced (<400 nm) Au electrodes. The 6T grains grow between the four contacts and their electrical behavior is probed as a function of temperature and gate voltage applied to the substrate. From these data, we extract the Au-6T contact resistance and determine its sensitivity to both gate field and temperature.

11:00 AM \*H6.3 METAL-ORGANIC INTERFACE INTERACTIONS. <u>LC. Feldman.\*\*</u> M. Huang.\*, M. Morales.\*, K. McDonald.\*, S. J. Rosenthal.\*, R. A. Weller.\*\* Dept of Physics and Astronomy. Vanderbilt University, Nashville TN. \*Dept of Chemistry, Vanderbilt University; \*Dept of Electrical Engineering, Vanderbilt University; \*Solid State Division, Oak Ridge National Lab., Oak Ridge, TN.

Interfaces formed by the combination of organic materials and metal contacts will undoubtedly prove to be a problematic aspect of employing molecular electronics. Much of the information on this important interface originates in the field of organic light emitting diodes (OLEDs), where the interplay between the structural aspects of the interface and the electronic aspects of the organic materials are revealed. This talk will focus on the metal/organic interactions encountered in systems designed for organic light emitting operation. Such systems may be models for the molecular electronic systems envisioned for the future. Structural studies, employing a high-resolution ion scattering apparatus, will be described for the aluminum/Alq systems. Evidence for metal-organic penetration, at very moderate temperatures, is correlated with the deteriorating

electronic/optical properties of the system. Our experiments, along with earlier reports, show that penetration can be curtailed through the use of an intermediate oxide, although the final device configuration must then be optimized with respect to the balance of carrier tunneling through the oxide and interface protection. Other examples of metal/organic interfaces will be reviewed and the impact of these materials studies with respect to molecular electronics will be

11:30 AM \*H6.4
ELECTRONIC STRUCTURE OF ORGANIC/METAL
INTERRACES STUDIED BY UPS AND KELVIN PROBE.
Kazuhiko Seki, Nagoya Univ, Res. Center for Materiala Science and
Dept of Chemistry, Nagoya, JAPAN; Hiroshi Oji, Eisuke Ito, Daisuke
Yoshimura, Naoki Hayashi, Yukio Ouchi, and Hisao Ishii, Nagoya
Univ, Dept of Chemistry, Nagoya, JAPAN.

Univ, Dept of Chemistry, Nagoya, JAPAN.

The interfaces of organic materials with other solids play important roles in the function of various organic devices such as organic light-emitting diodes (OLEDs), spectral sensitization in photography, organic solar ceils, and electrophotography. Also they should be important in future molecular devices, both in the central part of the device and at the connection with outside circuits. However, sensous experimental examination of such interfaces, as started only recently [1,2]. In this talk we focus our attention on the organic/metal interfaces, and summarize our understanding about (1) the energy level slignment right at the interface, and (2) possible band bending within an organic layer, mainly using the utechniques of UV photoemission spectroscopy (UPS) and Kelvin probe method [3]. As for (1), the formation of electric dipole layer was observed in most organic/metal interfaces, and its origin is discussed. As for (2), recent examination of the existence/absence of band bending in ultrahigh vacuum will be reported. It is also pointed out that there can be much effects of (1) atmosphere at sample preparation and measurements, and (2) chemistry and interdiffusion at the interface.

[1] K. Seki, H. Yanagi, Y. Kobayashi, T. Ohta, Phys. Rev. B49, 2760 (1994) (2) S. Narioka, H. Ishii, D. Yoshimura, M. Sei, Y. Ouchi, K. Seki, S. Hasagawa, T. Miyazaki, Y. Harima, K. Yamashita, Appl. Phys. Lett., 67, 1899 (1995), [3] For a recent review, H. Ishii, K. Sugiyama, E. Ito, K. Seki, Adv. Materials, 11, 605 (1999).

SESSION H7: Chair: James Murday Tuesday Afternoon, November 30, 1999 Cape Cod/Hyannis (M)

1:30 PM \*H7.1 CARBON NANOTUBE INTRAMOLECULAR DEVICES. <u>Zhen Yao</u>, Henk Postima and Cees Dekker, Dept of Applied Physics and DIMES, Delft Univ of Technology, THE NETHERLANDS.

Delft Univ of Technology, THE NETHERLANDS.

Single-wall carbon nanotubes promise to be ideal candidates for the ultimate miniaturization of electronic devices. Single-electron transistors employing metallic nanotubes and field-effect transistors employing semiconducting nanotubes have been demonstrated. We will focus on the more fascinating intramolecular devices. In particular, by introducing a single pentagon-heptagon topological defect pair into the hexagonal carbon bond network, two pieces of nanotubes with different atomic and electronic structures can be seamlessly fused together, thereby creating metal-metal, metal-semiconductor, or semiconductor-semiconductor junctions. We will show the electrical measurements of well-defined nanotube intramolecular junctions. A metal-semiconductor junction is found to act like a rectifying diode, that is, it exhibits nonlinear transport characteristics that are strongly saymmetric with respect to bias polarity. The conductance across a metal-metal junction appears to be strongly suppressed, which is consistent with tunnelling between the ends of two Luttinger liquids. We will also discuss transport measurements on intramolecular devices created by mechanical deformation.

2:00 PM \*HT.2 CARBON NANOTUBES: ELECTRICAL TRANSPORT PROPERTIES AND DEVICES. <u>Phaedon Avouris</u>, Richard Martel and Herbert R. Shes, IBM Research, T.J. Watson Research Center, Yorktown Heights, NY.

Single-walled nanotubes (SWNTs) are nearly ideal one-dimensional systems with unique electrical, mechanical, and thermal properties, which make them promising candidates for nanoelectronic device applications. We will discuss several aspects of nanotube behavior, transport properties, and device applications. We will show that strong nanotube-substrate interactions allow the manipulation, with scanning probe microscope (SPM) tips, of both position and shape on NTs at room temperature. Manipulation by SPM tips facilitates the

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measurement of the nanotube electrical properties and allows the fabrication of simple devices. An example of a field effect transistor (NT-FET) involving a single 1.5 nm diameter semiconducting nanotube as its channel will be discussed. This NT-FET behaves as a p-channel MOSFET, and the \$\overline{1}\$1 back-gate can modulate the current by 5 orders of magnitude. The high electrode-NT contact resistance acts as a tunnel barrier at low temperatures transforming the NT FET to a single electron transistor (SET). We will then discuss how one can produce SWNT rings from straight SWNTs, and how these rings can be used to elucidate the transport mechanisms in SWNTs taking advantage of electron interference effects. We will show that low temperature magneto-resistance measurements on the observation of the temperature dependence of the coherence length we determine that the dominant dephasing mechanism, at low temperatures, involves Nyquist electron-electron interactions, and that the SWNT ring is a state of weak localization in the temperature range of 3-60 K. At the same time strong electron correlation produces a Fermi level singularity Below about 1 K we observe a transition from the weakly localized to a strongly localized state where transport is thermally activated and, finally, below about 0.7 K a weak anti-localization behavior is seen.

2:30 PM \*H7.3

DEFECTS AND TRANSPORT IN CARBON NANOTUBES.

Steven G. Loug. Department of Physics, University of California at Berkeley, and Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA.

National Laboratory, Berkeley, CA.

The nanometer dimensions of the carbon nanotubes together with the peculiar electronic structure of a graphene sheet make these quasi-one-dimensional structures have highly unusual electronic properties. In this ralk, I will briefly review some theoretical work on the relation between the atomic structure and the electronic and transport properties of the carbon nanotubes. In particular, results on the quantum conductance of carbon nanotube junctions and tubes with defects will be presented. Calculations have been carried out using both ab initio and tight-binding methods. Metal-semiconductor, semiconductor-semiconductor, and metal-metal junctions have been studied. Other defects such as vacancies, substitutional impurities, and pentagon-heptagon defect pairs on tube walls are shown to produce interesting effects on the conductance. The effects of long-range vs. short-range disorders on the transport properties of metallic and electrostatically doped semiconducting carbon nanotubes are also examined, with the metallic tubes being much less affected by long-range disorders. This rich interplay between the atomic/structural properties and the electronic properties of the carbon nanotubes gives rise to new phenomena and the possibility of nanoscale device applications.

SESSION H8: Chair: Mark A. Ratner Tuesday Afternoon, November 30, 1999 Cape Cod/Hyannis (M)

3:30 PM H8.1
PHYSICS OF THE METAL-CARBON NANOTUBE INTERFACES:
CHARGE TRANSFERS, FERMI-LEVEL "PINNING" AND
APPLICATION TO THE SCANNING TUNNELING
SPECTROSCOPY. Yongqiang Xue, Supriyo Datta, Purdue
University, School of Electrical and Computer Engineering, West
Lafayette, IN.

Lafayette, IN.

Recent experiments on the single-wall carbon nanotube (SWNT) field-effect transistors and the scanning tunneling spectroscopy (STS) of supported SWNT show distinct behavior regarding the interface Fermi-level positionia. This raises the important question of whether the Fermi-level positionia depends on the contact geometry and/or the interface coupling. In this work we present a theory of the scanning tunneling spectroscopy of a single-wall carbon nanotube supported on the Au(111) substrate. The main results of our work are: (1) the work function difference between the gold substrate and the nanotube leads to charge transfers across the interface, which induce a local electrostatic potential perturbation on the nanotube side. This atomic-scale interfacial potential perturbation shifts the energy levels of the nanotube relative to the gold Fermi-level, and gives rise to the observed Fermi-level shift in the STS current-voltage characteristics. (2) for transport in the direction parallel to the nanotube axis, as in the case of nanotube transistors, the local potential perturbation aft the interface is not important in determining the Fermi-level position if the coupling between the metal and the nanotube is strong (i.e., low resistance contact). Any discrepancy between the metal Fermi-level and the nanotube "charge-neutrality level" should be screened out rapidly by the metal-induced gap state (MIGS) in the nanotube side,

leading to the "pinning" of the Fermi-level around the mid-gap position at low temperature. (3) we have taken the localized 5d orbitals of the platinum tip into account in our treatment of the STS which are responsible for the atomic resolution imaging of the nanotube.

3:45 PM H8.2 SWNT BASED CIRCUIT: THE DOUBLE QUANTUM DOT. J. Lefebvrg. M. Radosavlevic, A.T. Johnson, Dept of Physics and Astronomy and LRSM, University of Pennsylvania, Philadelphia, PA.

We present data obtained on multiple SWNTs (single-wall carbon nanotubes) circuits. In particular, SWNTs put on top of one another behave as two quantum dots in series. Calculations show that the junction is responsible for the tunnel barrier between the two dots. The tunneling amplitude of the barrier can be tuned electrostatically

4:00 PM \*H8.3 QUANTUM TRANSPORT AND DATA STORAGE IN NANOTUBE DEVICES. <u>David Tománek</u>, Department of Physics and Astronomy, Michigan State University, East Lansing, MI.

DEVICES. David Tománek, Department of Physics and Astronomy, Michigan State University, Beat Lansing, MI.

Carbon nanotubes and related fullerene structures are investigated for their usefulness as memory and logic elements, as well as manipulation tools, in molecular electronics. Multi-wall carbon nano-capsules containing charged fullerene ions, such as KaCto, behave as tunable two-level systems. Transitions between the two states can be induced by applying an electric field between the end caps of the outer capsule. Since the position of the encapsulated fullerene can be associated with a bit value, these systems can be used as nonvolatile memory elements [1]. Due to their small size and absence of incoherent scattering, nanotubes themselves behave as quantum conductors. Whereas the conductance of ballistic conductors is an integer multiple of the conductance quantum in most systems, the weak inter-wall interactions in multi-wall nanotubes may block some of the quantum conductance channels and redistribute the current non-uniformly over the individual tubes. These results provide a natural explanation for the unexpected non-integer conductance values observed for multi-wall nanotubes [2].

Nanotubes may also be used for a precise, semi-continuous deposition of atoms on nanostructures. Such atoms, when intercalated in nanotubes are dragged by an electric current, which may be induced and controlled by the relative phase shift between two laser beams irradiating the tubes at frequencies w and 2w [3]. Supported by the Office of Naval Research under Grant Number N00014-99-1-0252. Young Kyun Kwon, David Tománek, and Sumio Iijima, "Bucky-Shuttle" Memory Device: Synthetic Approach and Molecular Dynamics Simulations, Phys. Rev. Lett. 82, 1470 (1999).

Stefano Sanvito, Young-Kyun Kwon, David Tománek, and Colin J. Lambert, Fractional quantum conductance in carbon nanotubes (aubmitted for publication).

Petr Král and David Tománek, Laser driven atomic pump, Phys. Rev. Lett. 82, 5373 (1999).

4:30 PM \*H.8.4 NOVEL BEHAVIOR IN NANOTUBE-JUNCTION DEVICES F. Léonard and J. Tersoff, IBM T.J. Watson Research Center, Yorktown Heights, NY.

Yorktown Heights, NY.

Carbon nanotubes (NTs) hold great promise for nanoscale electronic devices. Though only a nanometer across, they have exceptional strength and stability, and they can be either metallic or semiconducting. An exciting possibility lies in devices fabricated on a single tube, which in principle permit extremely small size and high density. In this talk, we present results on the properties of p-n junctions, n-i junctions, and Schottky barriers made on a single-wall carbon NT. We find that NT devices differ dramatically from classic planar bulk devices. In contrast to bulk junctions, the depletion width for NTs varies exponentially with inverse doping. In addition, there is a very long-range (logarithmic) tail in the charge distribution, extending over the entire tube. Another major difference is that, for typical metallic contacts to NTs, Fermi-level pinning is weak or nonexistent. Even when there is strong pinning, the barries seen in transport will be radically different from that expected based on the traditional picture of pinning. Our general conclusions should apply to a broad class of NT heterojunctions, and to other quasi-one-dimensional "molecular wire" devices.

SESSION H9 Chair: Ari Aviram

Wednesday Morning, December 1, 1999

Cape Cod/Hyannis (M)

8:30 AM <u>\*H9.1</u> THEORY OF ELECTRONIC TRANSPORT IN MOLECULAR



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JUNCTIONS. <u>S.N. Yaiiraki</u> and Mark A. Ratner, Dept. of Chemistry and Materials Research Center, Northwestern University, Evanston, IL.

Chemical synthesis, whether based on molecular self-assembly techniques or end atom modifications, combined with advances in measurement probes such as scanning probe microscopy or break junction techniques have made the measurement of current through single molecules or molecular strands possible. We study theoretically the electronic transport in such molecular wire circuits. We present a time-independent scattering formalism, based on the Landauer formula of mesoscopic physics, that includes a description of the molecule and the metallic contact electrodes. The molecular sawell as the molecule/metal coupling description is obtained from ab-initio methods. The conductance of junctions of experimentally relevant systems is obtained. Effects of chemical bonding, geometry, dimensionality and neighboring molecules in adlayers on the conductance are discussed. Our results show that the description of the isolated molecular system is not sufficient to capture the behavior of the whole junction. Finally, we comment on another determining factor for the conductance of these systems, namely the location of the Fermi energy of the metallic contact relative to the molecular energy levels. To test the prevailing assumption that the Fermi energy occurs at mid-gap, we study the electronic structure of the molecular energy levels. To test the prevailing assumption that the Fermi energy occurs at mid-gap, we study the electronic structure of the molecular energy levels. To test the prevailing assumption that the Fermi energy occurs at mid-gap, we study the electronic structure of the molecular energy levels in the molecular energy levels in the molecular evels to account find that the Fermi level lies closer to the molecular elected as to account for the molecule-metal interaction. Differences and similarities with dots, tubes and Luttinger liquids will be emphasized.

9:00 AM \*H9.2 MOLECULAR SCALE ELECTRONICS. <u>James M. Tour</u>, Department of Chemistry and Center for Nanoscale Science and Technology, Rice University, Houston, TX.

Synthetic organic routes to precisely defined conjugated macromolecules (molecular scale wires) will be described using solution and solid phase approaches. The molecular scale wires are based on poly(phenylene ethynylene)s and poly(thiophene ethynylene)s and they possess thiol, selenol, and tellurol end groups to function as molecular scale alligator clips. Several molecular based resonance tunneling diodes (RTD) have been demonstrated. Potential routes to molecular based CPUs will be outlined wherein electrostatic potentials are uses as the information-carrying packets. A route to overcoming the detailed need for nanolithography will be described using self-assembling molecular systems possessing logic and memory within a molecular-based CPU.

9:30 AM \*H9.3 SELF-ASSEMBLY OF NANOSCALE COMPONENTS FOR MOLECULAR ELECTRONICS. Theress S. Mayer, Thomas N. Jackson, Michael J. Natan, Thomas E. Mallouk, Department of Electrical Engineering, Pennsylvania State University, University

Park, PA

Nanoscale and molecular electronics promise to deliver ultra high-density memory and logic circuits that can be realized with dimensions well below the scaling limits of conventional microfabrication techniques. To realize this promise, considerable attention has been devoted to developing molecular-level devices that function as nonlinear circuit elements and nanowires that interconnect these circuit elements. In this talk, we will provide a review of recent research activities related to the fabrication and characterization of nanoscale components including segmented metal rods and diodes, and the development of techniques that can be used to assemble these components into two- and three-dimensional networks. In particular, we are using membrane replication methods to make high aspect ratio, segmented metal rods consisting of alternating layers of Au, Pt, Ag, and Pd that are well suited for orthogonal self-assembly strategies. We have observed using fluorescence microscopy of Au-Pt-Au tipped rods that the Au regions of the rods can be derivatized selectively relative to the Pt with chain-terminating groups such as amines, carboxylates, or single-strand DNA. Using a combination of field- and chemical-assisted assembly techniques, we have aligned and attached 100 200 nm diameter segmented rods with good uniformity and reproducibility onto templated SiO<sub>2</sub> substrates. This has allowed us to characterize electrically simple segmented metal rods as well as more complex rods containing active components at the interface between the metal layers.

SESSION H10: Chair: Mildred S. Dresselhaus Wednesday Morning, December 1, 1999 Cape Cod/Hyannis (M)

10:30 AM H10.1 SYNTHESIS AND CHARACTERIZATION OF CONJUGATED MOLECULAR WIRES THREADED BY INSULATING TUBULES: Intae Kim, Yadong Yin, Younan Xis, University of Washington, Department of Chemistry, Seattle, WA.

We have demonstrated a self-assembly approach for the synthesis of conjugated organic polymers threaded by molecular tubules, with poly(p-phenylenevinylene) (PPV) and cyclodextrin rings as the example. In water, beta-cyclodextrin and stilbenoid compounds terminated in sulphonium end groups were self-assembled (as driven by hydrophobic interactions) into stable inclusion complexes; these complexes were subsequently polymerized into precursor polymers to PPV by addition of NaOH. Finally, the assynthesized precursor polymers were converted into conjugated polymer PPV by thermal elimination of sulphonium groups. The potential use of this new type of molecular wires will also be discussed.

10:45 AM <u>H10:2</u>
END-GRAFTED SEMICONDUCTING POLYMER—CANDIDATE
FOR MOLECULAR WIRE. <u>Kazuski Furukawa</u>, Keisuke Ebata,
Michiya Fujiki, NTT Basic Research Laboratories, Kanagawa,
JAPAN.

Molecular wire, a nanoacale lead, is attracting considerable attention not only for key technology to realize molecular devices in the next century but also for basic research to reveal intrinsic optoelectronic properties at the single molecular level. For the macromolecules directed to molecular wire use, perceptualists as re(1) conductivity, (2) connectivity, and (3) rigidity. Here we report on polysianes, a synthetically accessible silicon-catenated polymer with such semiconducting properties as photoconductivity, high hole drift mobility, and electroluminescence, as a candidate for a molecular wire. We demonstrate an end-grafted rodlike polysiane, covalently bonded not a Si(11) surface at one end and stretched more than 500 mm in length on the surface. We developed a unique "end-graft" technique to fix individual polysialne molecules homogeneously and extensively with a controllable density on a substrate surface. This was achieved by chemical reaction of between a reactive anchor of alkylbromide built on the surface and an end-lithiated polysiane. The substrate was prepared by photochemical reaction of BF(CH<sub>2</sub>)<sub>2</sub>CH=CH<sub>2</sub> with Si-H on an atomically flat SiH(111) surface. The latter was prepared by either living anion polymerization of masked disilene or scission reaction of the polysiane by a lithium reagent. Polysialne sar sunique in that the backbone rigidity is extensively varied depending on the steric effects of the organic substituents. The persistence length of rigid polysialnes reaches ca. 90 mm in solution at room temperature (longer than DNA ~60 mm!). We applied the above "end-graft" technique to the rodlike polysiane as "macromolecular roopes" lying on the surface. Because the density of the end-grafted polysiane is controllable by using substrates with different reactive anchor densities, we can place single polysiane molecules in a desired area, for instance between electrodes. A project to measure the LV characteristics of end-grafted polysianes us now in progress.

11:00 AM H10.3
FABRICATION AND TRANSPORT PROPERTIES OF Te-DOPED BI NANOWIRE ARRAYS. Y.M. Lin, Dept. of Electrical Engineering and Computer Science; X. Sun, S.B. Cronin, Dept. of Physics; M.S. Dresselhaus, Dept. of Electrical Engineering and Computer Science, Dept. of Physics; J.Y. Ying, Dept. of Chemical Engineering, Massachusetts Institute of Technology, Cambridge, MA.

Massachusetts Institute of Technology, Cambridge, MA.

Bi nanowire arrays with wire diameters ranging from 10 nm to 120
nm have been fabricated by pressure injection of liquid bismuth into
porous anodic alumins template. Because of the small electron
effective mass and the highly anisotropic Fermi surface of Bi, the
nanowire system is predicted to be a promising material for the study
of the quasi one-dimensional (1D) electronic system and possibly for
hermoelectric applications. The contributions from electrons and
holes can be decoupled by altering the Fermi level so that the
nanowire system becomes either n-type or p-type. Using similar
fabrication techniques, Te-doped Bi nanowires with different doping
concentrations are prepared providing a n-type nanowire system. The
XRD study of the doped nanowire arrays shows that the crystallinity
of the bulk Bi is preserved and the wires exhibit a preferred (101)
orientation. The semimetal-semiconductor transition of the nanowires
due to quantum confinement for this wire orientation is calculated to
occur at a diameter of 49 nm. In this paper, we report the
dependences of the transport properties, used as electrical resistance,
magnetoresistance on temperature, wire diameter and the doping
concentration. Based on the band structure of the bulk Bi and the
2-band Lax model, the theoretical transport properties of the quasi
1D system for various carrier concentrations are calculated and
compared with the experimental results. Preliminary experimental

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results of the Seebeck coefficient will also be discussed. We gratefully acknowledge the support of MURI subcontract 0205-G-7A114-01, NSF grant CTS-9257223 and DMR-98-04734, and the US Navy contract N00167-92-K0082.

11:15 AM H10.4
TRANSPORT MEASUREMENTS OF INDIVIDUAL BI
NANOWIRES <u>5.B. Cronin</u>, Y.M. Lin, X. Sun, Z. Zhang, J.Y.
Ying, M.S. Dresselhaus, Department of Physics, Department of Electrical Engineering and Computer Science, and Department of Chemical Engineering, Massachusetts Institute of Technology, Cambridge, MA.

Cambridge, MA.

The extremely small effective mass of Bi makes Bi nanowires an interesting system for studying quantum behavior. Determination of the electronic transport properties of Bi nanowires using a 4 point measurement method is essential in understanding the electronic structure of these nanowires. The absolute resistivity attained by this 4 point method is also important in evaluating the potential value of Bi nanowires for thermoelectric applications. The Bi nanowires used in this work are single crystal with nearly the same crystal structure and lattice constant as bulk Bi. The Bi nanowires are prepared by nonlithographic means (self-assembly), by first filling a porous alumina template with molten Bi or with Bi vapor and then dissolving the template leaving a solution of free standing wires. Individual wires from the solution are then used for the I-V measurement. For practical applications it may be desirable to leave the wires in the template, however in order to perform a 4 point measurement it is necessary to remove the wires from the template. The technique and methodology for affixing four electrodes on a single free standing Bi nanowire are discussed in detail. Attention will be given to various difficulties in the experiment such as oxidation of the Bi nanowires, unfavorable mechanical properties of Bi, such as low melting point, and static discharge. The nanowires studied in this work range from 70 to 200 nm in diameter. We plan to extend the measurements to wires of much smaller diameters. Theoretical predictions of quantum effects on the electronic structure and transport properties of Bi nanowires will be given. We gratefully schowledge the support of MURI subcontract 0205-G-7A114-01, NSF grants CTS-925723 and DMR-9400334, and the US Navy contract N00167-92-K0052.

11:30 AM H10.5
PLASMONICS: ELECTROMAGNETIC ENERGY TRANSFER AND SWITCHING IN NANOPARTICLE CHAIN ARRAYS BELOW THE DIFFRACTION LIMIT. M.L. Brongersma, J.W. Hartman, and H.A. Atwater, Thomas J. Watson Laboratory of Applied Physics, California Institute of Technology, Pasadena, CA.

Atwater, I homas J. Watson Laboratory of Applied Physics, California institute of Technology, Pasadena, CA.

Integrated optics faces a fundamental problem in guiding, switching, and amplification of light in that structures must have dimensions comparable to the wavelength of the guided light. Recently, it was theoretically shown that this problem might be circumvented by transporting electromagnetic energy slong linear chains of metal nanoparticles below the optical diffraction limit. Our models show that this transport is coherent and relies on the near-field electrodynamic interaction between closely spaced nanoparticles that sets up coupled plasmon modes in the chain. We have modeled power flow through such "plasmon wires" and three-terminal "plasmon switches" consisting of linear chains and tee-junctions or crosses. These structures could form the protocypical circuit building blocks of a nanoparticle array of complex architecture. The energy dispersion relations for linear chains have been computed; two branches are found corresponding to the transverse and longitudinal modes of the chain. The propagation westeingth, bandwidths and group velocities can be estimated as functions of metal particle size, interparticle distance, polarization, and the dielectric properties of the host matrix. Model calculations were performed for 25 mm Ag nanoparticles in dielectric host materials such that the plasmon energy is  $\hbar u_p < E_p$ , the host material bandgap. It was shown that the group velocity in linear chains was  $u_p \approx 0.01$  to fer 50 mm center-to-center particle spacing and the 1/p propagation was shown that the group velocity in linear chains was  $u_p \approx 0.01$  to fer 50 mm center-to-center particle spacing and the 1/p propagation bength was 500 mm implying the possibility of energy transport over several microns. Plasmon witches are structures in which the power flow through a plasmon wire can be modulated at a te-ejunction by changes in the field amplitudes or the input polarization in the "control wire". We

11:45 AM H10.6
FABRICATION AND CHARACTERIZATION OF EXTREMELY
NARROW METALLIC WIRES. D. Natelson, R.L. Willett, L.N.
Pfeiffer, K.W. West, Bell Laboratories, Lucent Technologies, Murray
Hill, NJ.

We have developed a general method for producing electrically continuous metal wires greater than one micron in length with wid below 5 nm, the size scale of multiwalled carbon nanotubes. These

wires can be made singly or in arrays, from a variety of materials, and should allow us to study normal metals, magnetic materials, magnetic multilayers, and superconductors in a previously inaccessible size regime. We present preliminary conductance measurements on normal metal wires of several diameters less than 20 nm, from 300 K to below 100 mK in fields up to 8 Tesla.

SESSION H11/G6: JOINT SESSION:
NANO TO MOLECULAR SCALE ELECTRONICS OF
ORGANIZED STRUCTURES
Chairs: Samson A. Jenckhe and Jane Shaw
Wednesday Afternoon, December 1, 1999
Salon E (M)

1:30 PM H11.1/G6.1

NANOSCALE PATTERNS OF METAL NANOPARTICLES
CHEMICALLY-ASSEMBLED ON BIOMOLECULAR SCAFFOLDS:
ASSEMBLY STRUCTURE, STABILITY AND ELECTRON
TRANSPORT PROPERTIES. James E. Hutchison, Leif O. Brown,
Jana Mooster, Scott M. Reed, Mary E. Schmidt, Dept of Chemistry
and Materials Science Inst. Univ of Oregon, Eugene, OR; Laura I.
Clarke, Martin N. Wybourne, Dept of Physics and Astronomy,
Dartmouth College, Hanover, NH.

Dartmouth College, Hanover, NH.

The novel electronic properties (e.g., Coulomb blockade) of nanometer scale assemblies of metal nanoparticles make them potentially useful in nanoelectronic devices and extremely sensitive chemosensors. To date, no straightforward and reproducible methods are available for the fabrication of low-dimensional nanoparticle assemblies. The fabrication of low-dimensional nanoparticle assemblies, the fabrication of nanoscale lines is a particularly challenging, yet important goal. Our method for nanofabrication of linear nanoparticle arrays involves the assembly of functionalized metal nanoparticle arrays involves the assembly of functionalized metal nanoparticle arrays involves the casefolds cast upon an insulating substrate and bridged between narrowly spaced electrodes. Our previous investigations of unpatterned nanoparticle thin films provided clear evidence of Coulomb blockade at room temperature, but the response was unstable over time. In this paper we present a wet chemical approach to preparing one- and two-dimensional arrays of gold nanoparticles assembled onto a polypeptide (poly-L-lysine) scaffold layer. The assembly process provides a simple, chemical method to immobilize the particles and is found to stabilize the electrical response (Coulomb blockade) of the array compared with unpatterned samples. A striking feature of the electrical properties is that the electron transport properties are dominated by transport thought one-dimensional chains within the sample. Structural studies by AFM and XPS will be presented that support the transport findings and provide evidence for the formation of one-dimensional assemblies within the arrays.

1:45 PM H11.2/G6.2
CHARGE TRANSPORT IN SEMICONDUCTOR QUANTUM DOT SOLIDS. C.A. Leatherdale, N.Y. Morgan, I. Prasad, M.G. Bawendi, and M.A. Kastner, Departments of Chemistry and Physics, Massachusetts Institute of Technology, Cambridge, MA.

Massachusetts Institute of Technology, Cambridge, MA.

Close-packed arrays of semiconductor quantum dots represent a model system in which to study the evolution of electronic structure in an artificial solid. We study photoconductivity in close-packed solids of nearly monodisperse C68 semiconductor nanocrystals. We observe quantum dot size and surface passivation dependent photoconductivity that can be qualitatively understood by considering the energy required to overcome the Coulomb energy of the initial electron-hole pair. The temperature dependence of the photocurrent indicates that tunneling processes dominate both charge separation and charge transport. We model the initial charge separation step using simple tunneling calculations. Simultaneous fluorescence quenching and photoconductivity measurements suggest that many more charge separated pairs are created than reach the electrodes. Measurements of the fluorescence intensity as a function of charge in the film, reveal that excess charge causes reversible photodarkening of the quantum dot solid accounting for some of the discrepancy. Hysteretic behaviour and long time transients, observed in both the photoconductivity and conductivity measurements, suggest that charge is easily trapped in the solid. Strong electron-electron interactions between charges on different sites in the lattice may be limiting the carrier mobility.

2:00 PM H11.3/G6.3

OPTICAL PROPERTIES OF SEMICONDUCTOR QUANTUM-DOT PHOTONIC CRYSTALS. Yu. A. Vlasov, M. Deutsch and D.J. Norris, NEC Research Institute, Princeton, NJ.

We explore the optical properties of semiconductor quantum-dot photonic crystals. These materials are made by combining colloidal chemistry with two steps of hierarchical self-assembly. First,



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monodisperse sub-micron silica spheres slowly settle onto a flat substrate and self-organize as a face-centered cubic lattice that is periodic on an optical length scale. Second, this template serves as a three-dimensional scaffolding for the self-assembly of colloidal CdSe nanocrystals into densely packed arrays, referred to as quantum-dot solids. Subsequently, the silica template can be removed by selective etching and a three-dimensionally patterned material consisting solely of semiconductor quantum dots sobtained. The resulting material is particularly interesting as a photonic crystal since we control several key parameters that determine its final behavior. First, we control the unique optical properties of individual semiconductor quantum dots. They have discrete optical transitions, efficient luminescence, high gain and optical nonlinearities, which are tunable with the size of the nanocrystal. Second, we control the structure of the silica spheres, we determine how the quantum-dot solid is patterned on an optical-length scale. This structure modifies the internal electromagnetic environment and influences the propagation of photons through the material. Here we study the optical properties of such semiconductor quantum-dot photonic crystals by transmission, reflection and diffraction. We present results that show the strong influence of the periodic structure of the photonic crystal on the photodiuminescence of the semiconductor quantum-dot solid.

2:15 PM H11.4/G6.4

2:15 PM H11.4/G6.4
SYNTHESIS OF LUMINESCENT ULTRA-SMALL SI NANO
PARTICLE COLLOIDS AND THIN FILMS. Munir H. Nayleh, J.
Therrien, O. Akcakir, G. Belomoin, Z. Yamani, N. Barry, E. Grattor
University of Illinois at Urbana-Champaign, Department of Physics,
Urbana, IL.

We describe a procedure for conversion of solid porous silicon into a dispersed colloid of ultra-small (~1 nm) silicon nano particles. When the colloid is excited at 355 nm, blue emission is observable with the naked eye, in room light. The luminescence is dominated by an extremely strong deep blue band at 390 nm, with a structureless weak corange/red tail, and a weak infrared band at 760 nm. We recorded, using two-photon infrared femto second excitation, the auto-correlation of the blue luminescence of a few particles in the laser interaction volume (of one pico cubic centimeter) including single particles. The measurements yield a particle's diffusion, size, and excitation efficiency comparable to those of fluorescein dye molecules one of the top efficient dyes in the blue. Thin films of particles are formed on device quality silicon substrates by gentle evaporation from a volatile (acetone) particle colloid, without compromising the blue emission. Two-terminal I-V spectra of the deposited film, taken using a scanning tunneling microscope, will be presented.

2:30 PM H11.5/G6.5 HIGHER-ÖRDRER SYNTHESIS OF MOLECULAR MAGNETS. <u>Sebastien Vaucher</u> and Stephen Mann, School of Chemistry, University of Bristol, Bristol, UNITED KINGDOM.

A major goal in contemporary materials science is the synthesis of molecular based materials that exhibit spontaneous magnetisation. Previously research approaches have been focused on the synthesis of new types of compounds, essentially by engineering at the unit cell level. A wide range of synthetic pathways to molecular magnets with appealing properties (high Tc, photoinduced magnetisation, etc.) are now available. Whereas it has been shown that inorganic materials, such as slites or titania, can be synthesised across a range of length scales by constructional, morphological and hierarchical coding of precipitation reactions, the analogous control of the higher-order structural features of molecular magnets has to our knowledge not been investigated. In this presentation, we illustrate how it is possible to use similar approaches to control the growth and organization of molecular magnets at the nano- and mesoscale. Several synthetic methods have been investigated and the products characterised by SEM and TEM. The general application of higher-order synthesis of functional molecular systems will be discussed.

2:45 PM H11.6/G6.6

ELECTROCHEMICAL METAL DEPOSITION CONTROLLED BY SELF-ASSEMBLED MONOLAYERS AND POLYMERS.
Andrea D. Weils, John T. McDevitt, William R. Murray, University of Texas at Austin, Chemistry & Biochemistry Dept, Austin, TX.

To more effectively process materials, new procedures were developed to anchor molecular reagents atop samples using self-assembled monolayers (SAM's) and polymers. Just as end group functionality allows specific ligands to interact with charged metal centers, appropriate choice of terminal functional group allows certain organics to interact strongly with metallic ions in the electrolyte solution. Through this interaction, organic layers can exert control over the electrochemical deposition of metal layers on high-Tc superconductors. Nucleation and growth of metal deposits on the surface are monitored by various electrochemical techniques. Electron

3:30 PM \*H11.7/G6.7 NANOSCALE PROCESSING OF ELECTROACTIVE MATERIALS. Michael Rubner, Department of Materials Science and Engineering, MIT, Cambridge, MA.

Nanoscale processing techniques have been utilized to manipulate a variety of electroactive materials such as light emitting Ru(II) complexes, inorganic nanoparticles and conjugated polymers into th film devices. Control at the nanoscale level has made it possible to dramatically improve the performance of these thin films. Details concerning the processing and electrical/optical properties of these new nano-composites will be discussed.

4:00 PM H11.8/G6.8

LAYER-BY-LAYER ASSEMBLY OF NANOPARTICLES. Arif
Mamedov, John Ostrander, Oklahoma State University, Chemistry
Department, Stillwater, OK, Parkhad Aliev, Departamento de Piaca
de la Materia Condensada, Universidad Autonoma de Madrid, Mario,
SPAIN; Miguel Correa-Duarte, Luia Liz-Marzan, Departamento de
Quimica Faisica, Universidade de Vigo, Vigo, SPAIN; Nicholas Kotov,
Oklahoma State University, Chemistry Department, Stillwater, OK.

Okishoma State University, Chemistry Department, Stillwater, OK.

The layer-by-layer assembly (LBL), i. e. the cyclic deposition of monolayers of oppositely charged materials, has been applied to the preparation of hybrid polyelectrolyte/nanoparticle thin films. This technique affords combining mechanical properties of polymers and unique physical characteristics of size-quantized clusters. Other advantages of the LBL processing of nanoparticulate films include simplicity, universality, tolerance to the substrate's shape, accurate control over the size, size distribution and structure of nanoparticles being assembled. Selection of nanoparticles defines the properties and the area of application of the produced assemblies. LBL films of CdS, edds, magnetite, yttrum iron garnet (YIG), cobait nanoparticles, and alumosilicate sheets yielded coatings with interesting light-emitting, electrical, magnetic, magnetoresistive, and magnetoopical properties. Importantly, the LBL technique opens a possibility to design sophisticated stratified structures. In particularly, the layer sequence was demonstrated to affect the coercivity, ion-sieving effect, magnetocoptical properties of magnetite, alumosilicate/polyelectrolyte, CdSe/alumosilicate/polyelectrolyte, and YIG/polyelectrolyte. CdSe/alumosilicate/polyelectrolyte, assemblies respectively. The LBL deposition was also applied to the preparation of films from naked and silica-coated nanoparticles of magnetite. The insulating SiO<sub>2</sub> costing affords isolation of individual magnetic grains and prevention of the exchange coupling between the nanoparticles. It was shown that the assembly of coated particles has a potential to reduce the noise of magnetic storage media and increase its storage capacity.

4:15 PM H11.9/G6.9
PHOTOVOLTAIC RESPONSES IN POLYMER-FULLERENE IONICALLY SELF-ASSEMBLED NANOSTRUCTURES.
PJ. Neyman, W. Graupner, J.R. Heflin, Virginia Tech, Dept of Physics, Blacksburg, VA; D. Marciu, M. Miller, A. Drake, Luna Innovations, Inc., Blacksburg, VA; H. Wang, H.W. Gibeon, H.C. Dorn, Virginia Tech, Dept of Chemistry, Blacksburg, VA; R.M. Davis, Virginia Tech, Dept of Chemical Engineering, Blacksburg, VA.

Virginia Tech, Dept of Chemical Engineering, Blacksburg, VA.

The ultrafast photoinduced electron transfer from conjugated polymers to fullerenes has allowed the production of efficient organic photovoltaic devices. Since the exciton diffusion distance is on the order of 10 nm, however, the charge transfer cannot occur unless the fullerene acceptor is within this distance of any given optically-excited electron-hole pair. We have recently fabricated polymeric photovoltaic devices from ionically self-assembled monolayers (1SAMs) and variations thereon. The ISAM nanostructure fabrication method simply involves the alternate dipping of a charged substrate into aqueous cationic and anionic solutions at room temperature. Each monolayer is fully formed within a couple minutes of immersion, and the bilayer thickness can be controlled from 0.3 to greater than 5.0 m by variation of the solution parameters. Large-arac, conformal, flexible, optoelectronic thin films can thus be fabricated with detailed with ease of fabrication and low cost. We have employed several approaches to combine the tetrahydrothiophenium precursor of poly(para-pheneylene vinylene) (PPV) with fullerenes. As two examples, hydroxylated Cap forms direct ionic bonds with the cationic PPV precursor while pristine Cap forms covalent bonds with the amine groups of a second polycation incorporated into the ISAM films. Comparison of the short-circuit currents, open-circuit voltages, and energy conversion efficiencies are presented for photovoltaic devices made from these and other novel polymer-fullerene self-assembled nanostructures.

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4:30 PM H11.10/G6.10
CHARGE TRANSPORT AND ELECTROLUMINESCENCE IN
NOVEL HETEROCYCLIC LIQUID CRYSTALS. Rong Fan, George
G. Malliaras, Cornell University, Dept. of Materials Science and
Engineering, Ithaca, NY; L. Sukhomlinova, S. Gu, R.J. Twieg, Kent
State University, Chemistry Dept., Kent, OH.

We have investigated charge transport in a family of novel liquid crystals containing five member ring heterocycles (oxadiazoles, thiadiazoles, etc.). Efficient electron transport was observed in the smectic phase The electric field and temperature dependence of the electron mobility was studied. Apart from being able to simultaneously function as liquid crystals and charge transport agents, these materials are highly fluorescent, allowing the fabrication of organic light emitting diodes. Their electroluminescent properties in devices with various electrode materials were studied.

4:45 PM H11.11/G6.11 SOL-GEL SYNTHESIS AND NONLINEAR OPTICAL PROPERTY OF SILICA THIN FILMS DOPED WITH CYANINE DYE J OF SILICA THIN FILMS DOPED WITH CYANINE DVE J AGGREGATES. Takeash Watanabe, Keinuke Asai, Kenkichi Ishigure, The University of Tokyo, Dept of Quantum Engineering and Systems Science, Tokyo, JAPAN; Hao-Shen Zhou, Itaru Honma, Electrotechnical Laboratory, Tasukuba, JAPAN, Akihiro Mito, National Research Laboratory of Metrology, Tsukuba, JAPAN, Makoto Puruki, Oasmu Wada, FESTA Laboratory, Tsukuba, JAPAN, Satoshi Tstaura, Corporate Research Laboratories, Fuji Xerox Co. Ltd., JAPAN.

It is widely known that J aggregates formed by cyanine dyes show strong nonlinear optical properties. In order to apply them to practical devices such as an optical switching device, many J aggregate material morphologies have been suggested. However, since the J aggregates are unstable in many matrices, it has been difficult to realize such applications. We tried to embed cyanine dyes such a 1,1-detehyl-2,7-cyanine bromide into thin silica films by simple sol-gel processing and succeeded in making the dye molecules to form stable J aggregates in the films. The films were prepared in the following way; tetraethylorthosilicate was mixed with ethanol and stirred. The solution was added with dilute HCI solution and stirred. And then, the dye was added to it and the sol-solution was stirred. Finally, the sol-solution was asted for finally, the sol-solution was stirred. Finally, the sol-solution was used for film deposition on glass substrates by spin casting. In spite of containing plenty of the J aggregates, these films are transparent, homogeneous in thickness and stable at room temperature. The J aggregation depends on both of the dye concentration in the sol-solutions and the spinning speed during spin casting. The third order susceptibility |  $\chi^{(3)}$  | of the films ware irradiated with the beam at 77 K in vacuum environment (< 10<sup>-3</sup> Torr.) The |  $\chi^{(3)}$  | of the film doped with dye till saturated concentration is 5 × 10<sup>-7</sup> esu at on-resonant wavelength of 577 nm, which is larger than that of other organic materials. In addition, according to measurement of the response time r of the optical nonlinearity by pump-probe method with fa pulse laser at 577nm, it was found that the 7 is sub-ps. Thus, stability of the J aggregates, large  $\chi^{(3)}$  and fast r can be expected on the silica film doped with the

SESSION H12: Chair: Shashi P. Karna Thursday Morning, December 2, 1999 Cape Cod/Hyannis (M)

8:30 AM \*H12.1 CONTROLLING THE LOCAL ENVIRONMENT AND CHEMICAL STATES OF ELECTRONICALLY ACTIVE MOLECULES IN SELF-ASSEMBLED MONOLAYER STRUCTURES. D. Allara, T. Dunbar, A. Hooper, P. Weiss, M. Cygan, L. Bumm, A. Bross, Dept. of Chemistry, Pennsylvania State University, University Park, PA.

There is accelerating interest in harnessing the electronic properties of individual molecules for new generations of devices. Among the critical issues in making such devices are how to control the positioning of electronically active molecules and how to control their chemical and electronic states. This talk will discuss recent work directed towards the placement of electronically active molecules into defects in pre-assembled monolayers and on the characterization of the geometry and the local environment of the molecules. In addition, recent results will be discussed on the effects of metal atom deposition with respect to making contacts and to alteration of the electronic states (doping) of molecules.

9:00 AM H12.2 UNIMOLECULAR RECTIFICATION DOWN TO 105 K, AND 3PECTROSCOPY OF HEXADECYLQUINOLINIUM

TRICYANOQUINODIMETHANIDE. Robert M. Metzger, Laboratory for Molecular Electronics, Chemistry Department, The University of Alabama, Tuscaloosa, AL.

Alabama, Tuscaloosa, AL.

Hexadecylquinolium tricyanoquinodimethanide, 1, was first synthesized by Ashwell and co-workers for non-linear optical applications, and was studied by Sambles and co-workers for rectification in Langmui-Blodgett multilayers between dissimilar metal electrodes. We have recently confirmed that 1 is a one-molecule rectifier of electrical current, between Al electrodes, as a Langmuir-Blodgett monolayer [Metzger et al., J. Am. Chem. Soc. 119:10455 [1987]]. The rectification persists down to 105 K [Chen & Metzger, J. Phys. Chem. B103: 4407(1999)]. The dipole moment is large in the ground state (43 Debyes) but much smaller in the excited state (3 to 9 Debyes): this heips explain how the rectification works, in a slightly modifed Aviram-Ratner fashion. The spectroscopy (Vi-LV, NMR, FTIR, XFS) is consistent with a witterionic ground state and a much less polar excited state. [Baldwin et al., J. Phys. Chem. B103: 4269 (1999)]. Experiments at 4.2 K (to detect IETS) are in progress, and may be reported if successful.

9:15 AM H12.3

ELECTRONIC STRUCTURE AND RECTIFYING BEHAVIOR OF M/C<sub>16</sub>H<sub>33</sub>-Q<sub>3</sub>CNQ/M JUNCTIONS. C. Krzeminski, <u>G. Delerue</u>, G. Allan, D. Vuillaume, IEMN Dept. ISEN, Lille, FRANCE; R.M. Metzger, Univ. of Alabama, AL.

Metzger, Univ. of Alabama, AL.

A well-known paper of Aviram and Ratner [Chem.Phys.Lett. 29, 277
(1974)] proposed that a molecule which has a good electron donor and a good acceptor separated by a sigma-bonded bridge could act as a molecular rectifier. Inspired by this proposal, recent experimental papers [Metzger et al., JACS (1998); Vuillaume et al., Langmuir (1999)] have presented measurements of the electrical response of C12432-Q3CNQ contacted by two metallic electrodes which for several samples is characterized by a clear rectifying behavior. In this work, we model this structure using first principles density calculations and semi-empirical tight brinding calculations. We show that the electronic states closer to the gap of the molecule are fully delocalized in spite of the presence of a large permanent dipole: this effect results from the presence of the pi-bonded bridge between the donor and the acceptor. We calculate the electrical response of the system and we show that it is particularly important to perform a self-consistent calculation of the full structure, including the molecular layer and the electrodes. We obtain that the electrical rectification observed experimentally could come from an anisotropy of the electric field in the structure.

9:30 AM <u>H12.4</u> I-V MEASUREMENTS ON MOLECULAR SHEETS AND STACKS. <u>C. Daniel Flubie</u>, University of Minnesota, Dept of Chemical Engineering and Materials Science, Minneapolis, MN.

C. Daniel Frisbie, University of Minnesota, Dept of Chemical Engineering and Materials Science, Minnespolis, MN.

Our research group is performing several different types of current-voltage (I-V) measurements on sheets and stacks of organic molecules. The sheet and stack structures consist of extremely thin, vapor-deposited lamellar crystals of organic semiconductors or spontaneously adsorbed organic monolayers. In one approach, we use a conducting stomic force microscopy (AFM) probe as a positionable electrical contact to molecular sheets contacted by a fixed electrode at the other end. This configuration allows I-V measurements as a function of the probe-electrode separation, yielding sheet resistivity of single monolayers and an estimation of the organic-metal contact resistance. The I-V characteristics in these two-terminal measurements are generally extremely non-linear and can be interpreted in terms of both a charge injection barrier and space charge limited transport. Conducting AFM tips may also be used to probe vertical charge transport through self-assembled monolayers on gold, and the I-V traces in these measurements show some differences from recently reported STM studies on the same systems. In a second approach, we employ electron beam lithography to fabricate fixed source and drain contacts to molecular sheets grown on SiO<sub>2</sub>/Si substrates. We use the resulting metal/organic/metal structures in a transistor geometry to probe the field effect conductance of the sheet (the doped Si substrate serves as a gate electrode). These three-terminal measurements allow determination of the carrier mobility as a function of temperature (5-300 K) and the number of discrete molecular layers in the sheets. Importantly, we find metal-coated AFM tips may be used as potentiometric sensors to map voltage distributions in the molecular intest transport experiments. The potential maps show the distribution of AFM imaging with transport measurements, facilitating correlation of transport properties with specific, well

9:45 AM <u>H12.5</u> FABRICATION METHODS FOR GOLD NANOCLUSTER DEVICES: <u>A. Snow</u>, M.G. Ancona, W. Kruppa, D. Park, J.B. Boos



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and G.G. Jernigan, Naval Research Laboratory, Washington, DC.

and G.G. Jernigan, Naval Research Laboratory, Washington, DC. The possibility of chemically self-assembling nanoelectronic devices and circuits has attracted wide attention because of its potential simplicity and favorable economics. In this work, a hybrid approach is explored in which pre-patterned nanoscale gold electrodes guide the self-assembly of ligand-attabilized gold nanoclusters. The gold nanoclusters are composed of a gold core (1-5 nm dismeter) encapsulated by an alkanethiolate or phenethylthiolate monolayer (4-15nm thick). The deposition chemistry involves functionalizing the gold electrodes and/or the \$102\$ surface with chemical coupling agents. Clusters become immobilized on the surface when the surface-anchored coupling agents displace thiols in the ligand shell. XPS measurements were used to characterize the efficiency and selectivity of this deposition chemistry and to explore methods for enhancing its selectivity by surface passivation. By repeated immersions in cluster and dithiol solutions multiple cluster layers were deposited forming a uniform resistive coating. The start-up of the deposition is delayed (3 cycles) indicating a spacing of about 10 nm between the "seed" clusters initiating the deposition. With hexanethiol-coated clusters the sheet resistance is found to be \$2.10<sup>11</sup> (1)quare per nanocluster layer. Similar results obtained on macroscopic-sized samples suggest this number is truly characteristic of the film and is not due to contact effects. This value is also consistent with theoretical estimates. When the same experiment is performed with phenethylthiol-coated clusters the resistativity is significantly lower and appears limited by the dithiol contacts to the Au electrodes. Discreteness in this conductivity suggests that the numbers of contacts is small and that effects of single molecule conduction are being manifested. Another area of experimentation has been to define narrow lines of clusters between the electrodes using dithiol bridging or lithography. With the former te

SESSION H13: Chair: Robert M. Metzger Thursday Morning, December 2, 1999 Cape Cod/Hyannis (M)

10:30 AM \*H13.1 CONJUGATED ORGANIC "MOLECULAR WIRES" -CONDUCTION CHARACTERISTICS AND INCORPORATION INTO METAL AND METAL/SEMICONDUCTOR NANOSTRUCTURES. R. Reifenberger, Purdue University, W.

Lafayette IN.

Key advances in the molecular electronics arens will require a better understanding of the electronic conduction mechanism in organic molecules and the coupling of suitable molecules into local circuit nodes. It will also be necessary to interface these 'molecular circuit elements' with macroscopic structures. With these issues in mind, we will discuss recent experiments and theoretical models that provide some answers to these important questions,[1-6] The conduction properties of conjugated organic molecules will be reviewed and prototypical metal and metal/semiconductor nanostructures which exploit molecular conduction will be described.

[1]. M. Dorogi, et al., Phys. Rev. B52, 9071(1995) and R.P. Andres, et al., Science 272, 1323(1996).

[2]. M. Samanta, et al., Phys. Rev. B53, 7626(1996) and S. Datta, et al., Phys. Rev. Lett. 79, 2530(1997).

[3]. W. Tian, et al., Physica E 1, 304(1997).

[4]. W. Tian, et al., J. Chem. Phys. 109, 2874(1998).

[5]. Y. Xue, et al., Phys. Rev. B58, 77852(1999).

[6]. T. Lee, et al., Appl. Phys. Lett. 74, 2869(1999).

This work was sponsored by the Army Research Office under Grant No. DAAL-03-92-G-0144 and NSF Grant No. 9708107-DMR.

11:00 AM <u>H13.2</u>
FORMATION OF Au<sub>35</sub> NANOWIRES AND HOLES BY ATOMIC FORCE MICROSCOPY. <u>R. Houbertz</u>, Institute of Experimental Physics, Saarbruecken, GERMANY and Sandia National Labs, Livermore, CA; U.E. Volmar, R. Steinkampf, Institute of Experimental Physics, Saarbruecken, GERMANY.

Atomic force microscopy (AFM) was used to generate Au<sub>35</sub> cluster wires from thick Au<sub>35</sub> cluster layers adsorbed on highly oriented pyrolitic graphite (HOPG) under ambient conditions. The wires were generated with and without applied bias. The extent of the resulting structures is dependent on the size of the applied bias. For zero or positive bias at the probe, the formation of cluster nanowires is observed, whereat for negative probe bias holes are generated. This observation is also confirmed by force- distance-curves. Particularly, the formation of holes cannot be understood by simply regarding and der Waals forces. It is shown, that the data can be understood in the framework of single-electron tunneling (SET) if a bias is applied.

Additionally, the interaction forces are calculated using a simple model. The calculated Coulomb forces between probe and sample are of the order of several hundred pN which is far above the acting van der Waals forces, thus being large enough to account for the generated

11:15 AM <u>H13.3</u>
THE FORMATION, OPTICAL AND ELECTROPHYSICAL
PROPERTIES OF THE POLYMER LANGMUIR BLODGETT
FILMS WITH LEAD AND MANGANESE SULPHIDES. Yu.N. Savin,
S.I. Gordeev, <u>A.V. Tolmachev</u> Institute for Single Crystals, Kharkov,
UKRAINE.

The optical and electrotransport properties of semiconductor nanoparticles in dielectric matrixes draw attention now because quantum dimension effects can be observed here and such structures can be used in photonics, micro- and nanoelectronics. The organized molecular assembles such as polymer Langmuir-Blodgett films evoke particular interest due to possibility to obtain the ordering molecular structures controlled in size. In this communication the investigation results of the formation processes of aromatic polyamic acid LB films, as well the nucleation and growth PbS and MnS nanoparticles in these films are reported. LB films containing semiconductor nanoparticles were obtained by two stages. At first, Langmuir monolayers of polyamic acid were formed at the subphase containing b(NO<sub>2</sub>); or MnSO<sub>4</sub> saits and then were transferred on the quartz and silicon substrates. At the second stage LB films were exposed in water solution of Na<sub>2</sub>S to obtain the nanoparticles of lead or manganese sulfides. Absorption of Pb-<sup>2</sup>F and Mn2<sup>2</sup>F ions on the Langmuir monolayers depending on pth of subphase are studied by XPS method. The nucleation and growth kinetics of nanoparticles depending on parameters determining the thermodynamics of polymer monolayer (pH, concentration of Pb, Mn and Sions, temperature, surface pressure) as well the duration of exposition in Na<sub>2</sub>S solution are investigated. It is found the spectral component in UV-VIZ absorption spectra is appeared after the exposition of LB films in Na<sub>2</sub>S solution. This component is due to nanoparticles of PbS or MnS in quantum-restricted state (Q-state). The effects of a temperature and duration of exposition in Na<sub>2</sub>S solution on the location of peak adsorption, long wave absorption edge and the nanoparticles sizes are studied. The photoconductivity and capacity-voltage characteristics are investigated versus the sizes and concentrations of a nanoparticles.

11:30 AM <u>H13.4</u>
ROOM TEMPERATURE SINGLE ELECTRON CHARGING OSCILLATIONS IN GOLD NANOPARTICLE NETWORKS FORMED UPON A BIOPOLYMER TEMPLATE. Laura I. Clarke, Martin N. Wybourne, Dept of Physics and Astronomy, Dartmouth College, Hanover, NH; Jana L. Mooster, Leif O. Brown, Scott M. Reed, and James E. Hutchison, Dept of Chemistry and Materials Science Institute, University of Oregon, Eugene, OR.

Science Institute, University of Oregon, Eugene, OR.

The past two decades have seen phenomenal growth in the study of artificially fabricated mesoscopic structures. Typically constraints imposed by the patterning technique have limited lateral feature size to around 20 nm. To prepare structures below this size, attention is being focused on intrinsic nanostructures, such as metal nanoparticles and single molecules. Metal nanoparticles that contain less than about one hundred atoms have a small enough inherent capacitance to make the charging energy at least an order of magnitude larger than the thermal energy at room temperature. While room temperature charging effects in single nanoparticles have been observed, a remaining challenge is to create arrays and architectures of metal nanoparticles that exhibit single electron charging persons. Although the electronic chains hat show single electron charging effects at room temperature. We have prepared networks of one-dimensional metal nanoparticle chains that show single electron charging effects at room temperature. We have prepared networks of one-dimensional metal nanoparticle arrays made by casting a film of the biopolymer poly-Llysine onto metal electrodes with subsequent exposure to 11-mercaptoundecanoic acid stabilized gold nanoparticles. The current voltage (L-V) characteristics of gold nanoparticles are promoted to the conductance is observed. We argue that these features arise from transport though one-dimensional regions of the network and show that the periodicity is consistent with the estimated capacitance of the nanoparticles. We will discuss the disorder in these s

11:45 AM H13.5 QUANTUM-DOT LEAD IODIDE FILMS FOR OPTICAL AND X-RAY IMAGING APPLICATIONS. <u>Krishna C. Mandal</u>, Bryce K. Dille, David Rauh, EIC Laboratories, Inc., Norwood, MA; O. Savadogo. Materials Engineering Department, Ecole Polytechnique, Montréal, Canada; Raghu N. Bhattacharya, National Renewable



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Energy Laboratory, Golden, Colorado; U. Sengupta, Jin Z. Zhang, Department of Chemistry, University of California, Santa Cruz, CA.

Department of Chemistry, University of California, Santa Cruz, CA.
This paper describes our recent research in developing large-area (4x4 eq. inch) lead iodide (Pb12) quantum dor (Q-dot) films for highly sensitive optical and X-ray imaging applications. The colloidal Pb12 nanocrystals were first synthesized in the solution phase reaction (80 v) ethanol) of Pb-based precursors in the temperature range 25-65 degree celcius. The synthesized ananocrystals were then isolated in sizes ranging from 12-120 Å in diameter and used for well-defined Q-dot film preparation. Free standing Q-dot Pb12 films of thickness approximately 25 micron were deposited on ITO-coated glass substrates by the colloidal spin-casting technique and were then thoroughly investigated by X-ray diffraction, electron probe microanalysis, transmission electrical resistivity, and charge transport property measurements. A large number of X-ray and photo-detectors were fabricated using lithographic masked metallization technique. The detection properties of signal amplitude for a given X-ray energy have been measured and was found to be about 6-8 times larger as compared to standard phosphor screens used for X-ray imaging. Charge transport characteristics and timing behavior of these nanocrystalline Q-dot films under bias operation have been measured, and the results demonstrated for the first time that these films are highly promising for real-time optical and X-ray imaging applications.



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