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14. ABSTRACT
Random boolean networks (RBNs) have been studied theoretically and computationally in order to be able to use their remarkable self-healing and large basins of attraction properties as quantum computing architectures, especially focused on problems of physical interest which do not require universal computational structures. This preliminary study was limited primarily to 1D strings, but eventually work should be directed beyond two state networks to multi-state ones. Available software wasused to study selected, important properties of RBNs in 1-D. The results of this preliminary study suggest that extensive work will be needed to apply the basic principles to physical manifestations, for example, building type II architectures in self-assembled spin lattices on a chip.

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Quantum Random Networks for Type 2 Quantum Computers

AFOSR Contract # FA9550 04-1-0409

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Project Dates: 06/01/04-05/31/06

Statement of Work:

Random boolean networks (RBNs) were to be studied theoretically and computationally in order to be able to use their remarkable self-healing and large basins of attraction properties as quantum computing architectures, especially focused on problems of physical interest which do not require universal computational structures. This preliminary study will be limited primarily to 1–D strings, but may go beyond two state networks to multi-state ones. Available software will be used to study selected, important properties of RBNs in 1-D. Eventually new codes will be created to go to 2-D nets as time and progress permit. The goal will be to determine the basic principles to enable physical manifestations, for example, building type II architectures in self-assembled spin lattices on a chip.

The specific questions to be explored were:
1. Finding a small class of update rules which lead to non-trivial basins of attraction whose number is small enough to handle.
2. Classifying these systems so as to find class 3 rules which are rich enough to do physical problems (pattern recognition, fluids etc.)
3. Create transition rules among basins of attraction, which is the analog of programming.
4. Find a method of implementing universal quantum gates
5. Find a method of coupling the gates to do some important physical problems, following the strategy of quantum lattice gases, which are known to work quite well.
6. Examine the impact of quantum decoherence on the system. The cases of fluid flow and pattern recognition can be done with small systems. Simulators would require at least 1000 x 1000 elements but would be astronomically fast.

This work was closely linked wherever possible with ongoing, separately funded, experimental work of the Allara group on physical implementation of q-bits and gates.

Progress towards achieving the research objectives:

The project work was initiated in October of 04. Between that time and 01/05 the theory approach was initiated. The software was set up on a PC platform with the idea of handling no more than 4 states to start. It was intended during the current period to increase to 6 but the complexity of the attractor basin structure as well as the space-time dynamics was too great to achieve this level so soon. The initial conclusions were that the basins are less random than thought and were organizing themselves into structures (*dogmas*) that were capable in principle of being handled as q-bits with gates added to allow computing.

Accomplishments/New Findings:

In the last progress report an overview of the project was given with indications of the new directions planned. During the final period the progress of the theory effort was unexpectedly hindered by personal health issues of Dr. Hasslacher who finally died in November 2005. After that time, the administrative PI worked with theory colleagues of Dr. Hasslacher to try and collect Hasslacher's ideas into a coherent framework and evaluate what possibilities there were for extension of the work by others. Also, computational resources were bolstered and efforts made to identify possible physical structures that could embody the RBN concepts.

With continued analysis of the problem, it became more and more evident that one needs a physical platform, even if very elementary, in order to make simple examinations of how arrays or swarms of interacting q-bits or classical bits interact with gates. For example, how a large interacting self-assembled spin array behaves with interacting q-bit structures. Allara continues to work under separate funding on fabricating or assembling arrays of self-assembled electron spin dots, both through pre-prepared stable arrays and through *in-situ* prepared arrays via chemical activation, that can serve as q-bits and on the reading and writing on these single spins using magnetic force resonance spectroscopy in collaboration with John Marohn at Cornell. This experimental approach, if successful, eventually would be a useful Type 2 platform.

We also looked into the feasibility of a platform of large arrays of nanofabricated nanostructures which can interact via near field interactions of electromagnetic plasmon resonance states. These arrays would be activated by near field injection of EM energy from a photon source with the proper momentum characteristics, and the flow of energy around the structure could potentially be used as an RBN type of system. Given that there are now advanced methods of molecular nanolithographies (Penn State is a center of excellence in this area) that can be used to fabricate EM confinement dot structures out of gold or silver, it may be very fruitful to examine candidate structures. In addition, recent mapping of EM wave behavior in confined geometries onto cosmological black hole behavior could prove to add a powerful tool to analyzing the behavior of these otherwise classical plasmon systems (e.g., see: *Nonlinear optics of surface plasmon toy black holes*, I. I. Smolyaninov, Xiv:cond-mat/030959 v1 25 Sep 2003; *Hawking Radiation in an Electromagnetic Waveguide?*, R. Schutzhold, and W.G. Unruh, *Phys.Rev.Lett*, 2005, *95*, 031301.)

Personnel Supported on this grant:

Faculty: David L. Allara, Penn State University (**no direct support, time/effort donated**)
Associated Professional Researchers: Dr. Brosl Hasslacher, with locations varying between Santa Fe, NM and the Center for Studies in Physics and Biology and the Laboratory for Mathematical Physics, Rockefeller University., Rockefeller University, NY, NY
Peer-Reviewed Publications: None
Interactions/Transitions: No transitions in this granting period.
New discoveries, inventions, or patent disclosures: None
Honors/Awards: Allara elected a Fellow of the AVS.

Attachment 1

Self-Assembled Molecular Spin Arrays for Computation

AFOSR Contract # F49620-01-1-0490

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Project Dates: 09/15/02-02/14/05

Statement of Work:

This project goal was to form a team of a theoretical physicist and a physical chemist to produce a molecule-based quantum logic system that can be scaled into a workable quantum computation system. The central ideal was to start with existing knowledge in QLG theory and the assembling of precision molecular films and couple the pieces to produce a precision molecular array of electron spins that demonstrates the general principles that can be followed for designing and building future QLG computing machines from molecular spin arrays.

Experiment:

- Development of new types of molecular arrays that contain a spin ½ electron in selected surface geometries
- Development of novel assembly methods that lead to highly organized monolayers with uniform intermolecular spacings and chemical stability
- Characterization of the arrays using a variety of advanced surface science tools including optical and infrared spectroscopy, atomic force microscopy, and various scattering methods

Theory:

- Perform theoretical analyses of the quantum properties of the qubit arrays in terms of developing topological models based on the quantum lattice gas approximation (QLGA). These analyses will be used to develop error correction codes and novel methods for quantum computing on the qubit arrays. Specific tasks include:
- Analysis of the role of noise/decoherence, including combined stochastic with unitary evolution to simulate the effect of incomplete experimental measurements, which will be the case for preliminary versions of the molecular array.
- Develop novel error correction methods for the qubit arrays including model decoherence and defects
- Develop QLGA algorithms for classes of nonlinear systems, including soliton systems
- Examine non-homogeneous operations that help approach a universal quantum gate
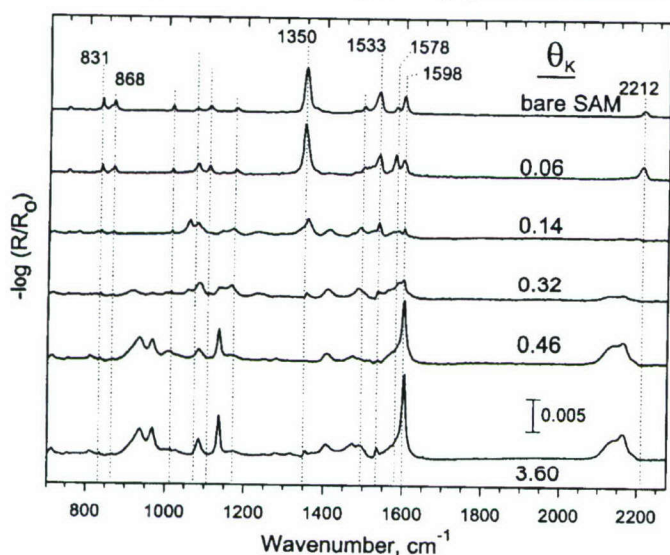
Project Results:

Experiment (Allara):

In the initial part of the project experiments were carried out to prepare a lattice of electron spins based on a self-assembled monolayer of a electron accepting molecules. In the first period we reported that a combination of quantitative infrared vibrational spectroscopy and density functional theory indicate that the addition of vapor alkali metal atoms to a self-assembled monolayer of a nitroaromatic molecule can create a spin lattice with ~1 nm spacings between the electron spins. This appeared to be the first example of a molecular spin lattice and work was done to characterize the molecular structures. In the later stages of the project the work was done in more detail and it was shown that complications with accurate dose measurements of the deposited vapor made the interpretations less straightforward than originally thought. Efforts also were made to build an apparatus for measuring sticking coefficients and to bring on line XPS and scanning probe capabilities for the experiments.

Initial Project Period:
Experiments involving dosing self-assembled monolayers of a fully conjugated nitroaromatic molecule with Li and K coupled with *in-situ* infrared reflection spectroscopy (IRS) were run. Using density functional theory (DFT) quantu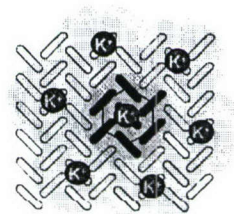m calculations to assist in interpreting the rather complex vibrational spectra, we concluded that both vapor deposited Li and K atoms can inject electrons into the molecules in the SAM and that type of state produced depends strongly on the alkali metal coverage. At

~ complete collapse of initial spectral features by 1 K per 7 SAM molecules

low coverages the added K appeared to add ~1 electron per 7 molecules, as illustrated in the figure above. This finding suggested the creation of a spin-containing center in one molecule surrounded by a 2-D shell of 6 nearest neighbor molecules and the charge residing in this shell induces repulsive interactions that do not allow further
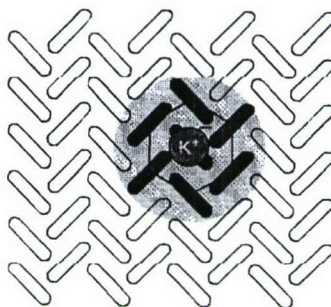
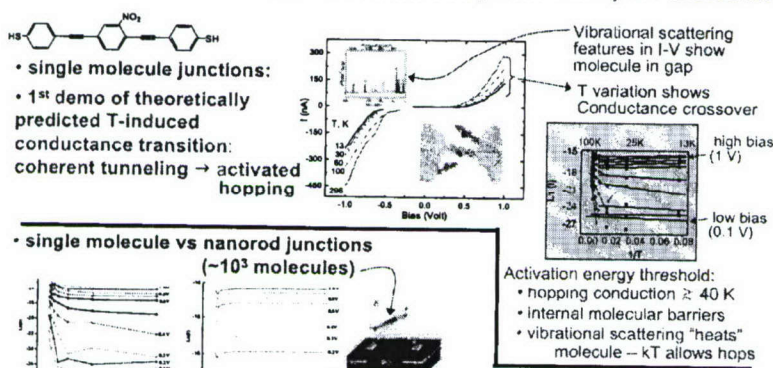e⁻ delocalized over NN shell??   Repulsive lattice?? (K/graphite)

charge injection until much higher metal atom coverages. We speculated that this structure is essentially a repulsive spin lattice, as illustrated in the schematic drawing. Overall these results demonstrated that electrons can be chemically injected into fully conjugated molecules in SAMs to form a variety of new types of charged molecular states with electron spin.

Interim Project Work on Electron Transport:

While waiting for new equipment to be brought up to operation, there was an opportunity to run a series of electron transfer experiments with electron acceptor molecules. In the case of a spin lattice one injects an electron into a stable state with a spin ½ with the charge pinned by the alkali metal counterion. The objective of the ancillary experiments was to bias the molecules at two sides and look at the charge transport rates of a electron that is injected (at a source electrode) but then transported to the second contact (drain electrode). The isolated junctions were made using electromigration gap formation in molecule covered, fabricated nanowires. The SAM junctions were made using Au nanorods with ~5000 molecules in the junctions. The exact same molecule was used in both types of junction to allow direct comparison. The results are summarized in the figure below.

First, with the isolated junctions the data show the onset of a hopping transition at increasing temperatures. This is the first measurement that shows the thermal effects on conductance mechanisms in molecular wires. Second, in contrast, the molecules embedded in a SAM show only coherent conduction at all temperatures. Further, in the coherent tunneling regime for both types of junctions as the bias approaches ~1 V, a single isolated molecule conducts over 10^3 times better than the average conductivity of a single molecule imbedded in a SAM. These results can be understood in terms of the screening effects of SAM molecules on the electrostatic potential drop across a bridge molecule and in the higher non-equilibrium temperature of

- single molecule junctions:
- 1st demo of theoretically predicted T-induced conductance transition: coherent tunneling → activated hopping

- single molecule vs nanorod junctions (~10^3 molecules)

Vibrational scattering features in I-V show molecule in gap

T variation shows Conductance crossover

Activation energy threshold:
- hopping conduction ≳ 40 K
- internal molecular barriers
- vibrational scattering "heats" molecule -- kT allows hops

Flat conductance T-dependence for 10^3 molecules vs one:
- molecular ordering freezes out onset of hopping transition in molecular ensembles
Coherent tunneling regime: 1 isolated molecule conducts >10^3 x 1 molecule in SAM
- isolated molecule: electron-phonon coupling + vibrational fluctuations causes higher conductance

Selzer, Cabassi, Mayer, Allara, *JACS*, 2004, *126*, 4052-4053
Selzer, Cabassi, Mayer, Allara. *Nanotechnology*. 2004. *15*. S483–S488

an isolated molecule compared to a SAM embedded molecule. These effects lead to enhancement of electron-phonon coupling and lowering of the HOMO-LUMO gap, both of which will increase the tunneling rates of an isolated bridge molecule.

Final Project Period:

Follow on efforts were made to characterize the physical and chemical nature of the spin lattice in more detail. At first it appeared that only one K atom was required to produce a spin delocalized cluster involving one ~7 molecules, viewed as possible a central molecule with the spin surrounded by a hexagon of 6 outer molecules. This picture was formed primarily on the stoichiometry of 1 K atom per ~7 molecules observed to cause complete loss of some of the

original SAM vibrational mode features in *in-situ* infrared spectra. Further work showed that the measurements of the K/molecule stoichiometry were more difficult than originally thought because of scattering of the K atoms off the monolayer and off the chamber walls. Direct experiments on the spin SAM were halted and the uhv infrared spectroscopy (IRS) system was redesigned over a year to work around this problem. In addition, in order to fully characterize the electronic state and any potential ordering in the spin SAM new experimental setups were designed and building started. Specifically, work was started to build a system to do *in-situ* X-ray photoemission spectroscopy (XPS) which could gather information on the electronic states of the spin molecules and to count the fraction of K atoms deposited that injected an electron into the molecules. Work also was started to build an in-situ AFM/STM which could image the surface of the spin SAM to look for critical patterns of the electronic states as well as image the initial ordering of the SAM prior to alkali metal deposition.

Once the new IR chamber was working better stoichiometry measurements could be obtained. The final results indicate that there is one K+ counterion associated with each molecular spin center so the lattice actually involves very localized spins rather than the possible case of delocalization over a shell of neighbor molecules as thought at one time.

These preliminary results with K (and Li) showed great potential for revealing a new class of molecular spin lattices but left several critical questions to be answered:
1. Do hexagonally ordered regions of uncompensated electron spin actually form on appropriate delocalized π-electron SAMs?
2. Do the sticking coefficients for alkali metals vary on these types of SAMs?
3. Since all applications of these SAMs to quantum computing would be at cryogenic temperatures, what is the effect of lowering T on the response of the SAMs to alkali metal deposition T.

In order to answer these questions we started on the new experimental capabilities in our lab:
1. Construction of a UHV chamber with *in-situ* AFM and STM capability to characterize the molecular resolution ordering of charges and metal atoms on ordered SAM surfaces at variable temperatures. Both the AFM and STM capabilities can be used to do molecular resolution imaging. Using the STM capability it may be possible to directly image the electron spins using the modulation of the STM current by the Larmor frequency of the unpaired electron in the vicinity of the STM tunneling current as shown in the diagram.
2. Construction of a precision quartz crystal oscillator microbalance (QCM) system for measurement of the sticking coefficients of alkali metals on SAMs.
3. Upgrading of the existing UHV IR chamber to do low temperature measurements
4. Construction of a new metal deposition capability in our existing X-ray photoelectron spectrometer system.
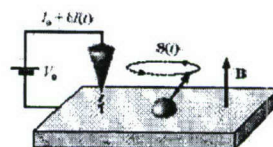
The QCM system was built and the first results showed that $S_K = 0.9-1.0$. This result indicated that the major problem with the K stoichiometry was due to wall and shield scattering. The

preliminary experiments were done also for Li, which showed a much lower sticking coefficient, $S_{Li} <0.25$.

The modifications to the existing XPS chamber to allow *in-situ* metal deposition were done using funds from other projects. Initial runs were started with K dosing of electron acceptor nitro aromatic SAMs. The experiments will be continued with other funding.

The uhv AFM setup was completed and running. Intial runs were started with K dosing of electron acceptor nitro aromatic SAMs. Of interest here are conducting probe AFM measurements of the spin and non-spin SAMs. The experiments will be continued with other funding.

The plans for the single spin STM were abandoned after learning from colleagues at Los Alamos National Labs that the experiment, as originally reported, did not work in practice because of amplifier noise that could not be defeated. However, plans are in progress for macro ESR runs to measure the presence and character of spins on alkali metal reduced SAMs. Vacuum transfer methods are now being designed to allow these experiments. This work will be continued with other funding.

Overall these experimental results demonstrate that electrons can be chemically injected into fully conjugated molecules in SAMs to form a variety of new types of charged molecular states with electron spin. These results further suggest that it may be possible to form an ordered lattice involving localized regions of ensembles of molecules bearing a spin at the center. This could be an important step in creating molecular spin lattices that offer new possibilities for consideration as spin arrays for quantum computing.

Theory (Hasslacher)

The last stages of the theory work were shifted to the new AFOSR project (Grant # F9550-04-1-0409) and for convenience the full results will be reported under that contract. Overall though, computer simulations started on simple Random Boolean Network systems of 4 states. The complexity of the attractor basin structure as well as the space-time dynamics has turned out to be astonishing. The basins of attraction are less random than expected and are organizing themselves into structures that we term *dogmas*. In principle, one could put q-bits and gates on them and develop a computational machine. Some of these ideas are summarized
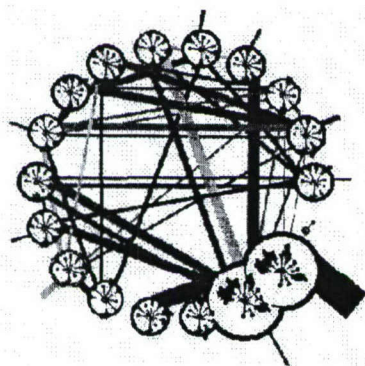
Essentially, when dealing with the possibility of an astronomical number of quantum spins that can be used as coherent state q-bits, the problem of unavoidable errors becomes a crucial one. Patches of defective spin arrays on assembly, after some time of operation, by natural chemical instability, or even by a defective algorithm and decoherence, all become sources of error. These error sources can't be corrected physically. It is also hard to map them because of the large numbers of elements involved. Before we spend considerable effort on generalizing lattice gas algorithms, it is important to know whether such error patches can be controlled and used as useful elements in themselves.

If we cannot control the place, type and extent of error patches, we may be able to control them as useful objects in themselves. This relies on their having a rich and useful dynamics. Error regions can then be used as powerful, even crucial q-bit objects.

We see that there is a spectrum of randomly organized networks emerging - some accidental and some intentional, ranging from defects in standard architectures, and going to the limit of random networks with an astronomical number of elements, which we call quantum "A" machines, after Avagadro's number.

By using a large number of random elements, we enter a regime in which statistical mechanics becomes valid, and we can use all the powerful tools developed there to help answer these questions. "A" machines belong to the class of random Boolean networks (RBNs). These have been studied by various groups over the last twenty years and are known to have fascinating and complex self-organizing properties which can be evolved and controlled.

When clocked and allowed to run from an initial global state, the behavior of these systems falls

naturally through transients into basins of attraction. These classify their phase or state space, a map of possible behaviors. The number and variety of these basins is quite large and tunable - they are very robust under defects in the network. A diagram of the network and basins is shown in the adjacent figure. The figure represents a transition diagram of how basins of attraction transit among one another in a typical set-up. The thickness of transition lines and color coding represent probabilities of transition. Controlling these is how one programs a RBN.

Each basin performs different functions ranging from logic to pattern recognition. They can be embedded in a quantum lattice gas algorithm, and could be made into novel quantum lattice gas machines. These attractors are not isolated - there is a transition matrix interconnecting them through which probabilities of transition from one basin to another can be adjusted by varying several parameters, as well as by levels of noise injected into the system. This is the analog of a finite state machine, although elementary operations may be more complex than simple quantum logic functions.

One can create a routing path that visits basins in a set order which can be changed dynamically. This can be realized by using a second network running at a slower clock rate which alters quantum transition rules locally and dynamically. So, a form of dynamic programming is available in the form of virtual quantum wiring of attractor basins.

In the final stage of the theory work more details were examined. The major accomplishment was the development of an overall conceptual approach to computing with Large Random Boolean Network (RBN) Systems. The main points are summarized in the figure. The overall idea is use classical or QC systems in the limit of huge numbers of elements. The networks can be randomly organized and can work by lattice gas rules. In this limit powerful phenomena come into play, in particular the use of second order phase transitions can provide powerful

computational aids via critical behavior and lead to universal computing machines.

Molecular-Scale (classical or QC) systems in limit of huge numbers of molecular components --- Deep Architecture + New Opportunities:
· Randomly organized networks (Avogadro machines) emerge -- use as RBNs:
- program in statistical mechanical (SM) behavior limit
- dynamically self-organizing, universal computation machines
- states governed by dynamic cellular automaton (or lattice gas) rules
- transient state flow into stable basins of attraction (BAs) robust to network defects
- different BAs specific to various problems, e.g., image recognition
- Interconnect BAs by parameter variation and noise injection
- Interconnect networks working at different clock rates --- dynamic alteration of transition rules can implement programming
- SM limit: novel phenomena (e.g., 2nd order phase transitions) arise to offer powerful computational aids via **criticality** and **universality**

Difference Greens function showing change in a BA under a simple 1-site change in the rule set. This is typical of the powerful tools available.
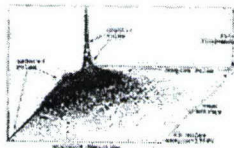
Summary:
huge molecular scale component network arrays offer possibility of astonishing, nonintuitive and powerful computational properties, both as classical and QC machines

Personnel Supported on this grant:
      Faculty: David L. Allara, Penn State University
      Associated Professional Researchers: Dr. Brosl Hasslacher, jointly in residence at the Center for Studies in Physics and Biology and the Laboratory for Mathematical Physics, Rockefeller University., Rockefeller University, NY, and in Santa Fe, NM.
      Postdoctoral Researchers: Drs. Yoram Selzer and Timothy Tighe, Penn State U.
      Graduate Students: none
Personnel Collaborating on this grant: Dr. Orlando Cabarcos, Professor Theresa Mayer, Penn State University,

Peer-Reviewed Publications:
1. *Nanoscale Molecular Switching of Thiol-Substituted Oligoanilines,* L. Cai, M.A. Cabassi, H. Yoon, O.M. Cabarcos, C.L. McGuiness, A.K. Flatt, D.L. Allara, J.M. Tour, and T.S. Mayer, submitted to *Nanoletters*
2. *Evolution of The Interface and Metal Film Morphology in The Vapor Deposition of Ti on Hexadecanethiolate Hydrocarbon Monolayers on Au,* Timothy Tighe, Tad Daniel, Zihua Zhu, Sundarajan Upilli, Nicholas Winograd and David L. Allara, submitted to *J.Phys.Chem.B*
3. *Chemical Pathways in the Interactions of Reactive Metal Atoms with Organic Surfaces: Vapor-Deposition of Ca and Ti on a Methoxy-Terminated Alkanethiolate Monolayer,* A. Walker, T. Tighe, B. Haynie, S. Uppili, N. Winograd and D. Allara, *J.Phys.Chem.B,* **2005** *109,* 11263-11272
4. *The Effect of Local Environment on Molecular Conduction: Isolated Molecule versus Self-Assembled Monolayer,* Y. Selzer, L. Cai, M.A. Cabassi, Y. Yao, J.M. Tour, T.S. Mayer and D.L. Allara, *Nanoletters,* **2005,** *5,* 61-65
5. *Temperature effects on conduction through a molecular junction,* Y. Selzer, M.A. Cabassi, T.S. Mayer, D.L. Allara, *Nanotechnology,* **2004,** *15,* S483–S488
6. *The Interaction of Vapor-Deposited Ti and Au with Molecular Wires ,* A.V. Walker, T.B. Tighe, J. Stapleton, B.C. Haynie, S. Upilli, D.L. Allara, N. Winograd, *Appl.Phys.Lett.,* **2004,** *84,* 4008
7. *The Dynamics of Noble Metal Penetration Through Methoxy-Terminated Organic Monolayers,* A.V. Walker, T.B. Tighe, O. Cabarcos, M.D. Reinard, S. Uppili, B.C. Haynie, N. Winograd and D. L. Allara, *JACS,* **2004,** *126,* 3954-3963
8. *Thermally Activated Conduction in Molecular Junctions,* Y. Selzer, M.A. Cabassi, T.S. Mayer, and D.L. Allara, *JACS.,* **2004,** *126,* 4052-4053

Interactions/Transitions: Several extremely significant and related discoveries on solid state, two-contact devices for probing temperature dependent current-voltage probes were done in collaboration with ongoing work funded by DARPA and the results were utilized by other workers in the DARPA Moletronics program.

New discoveries, inventions, or patent disclosures: Discoveries as noted above.

Honors/Awards (DLA):
- American Chemical Society, Adamson Award for Distinguished Achievements in Surface Chemistry (2003)
- Honorary Doctorate of Science, May 2003, Linkoping University, Sweden