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Theory and modeling in nanoscience: Report of the May 10-11, 2002 Workshop

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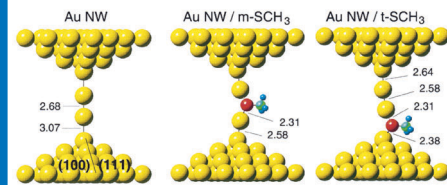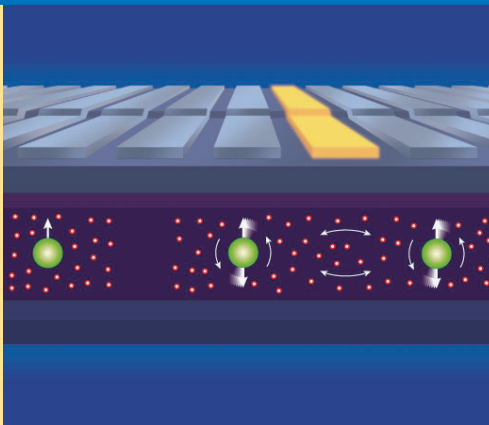McCurdy, C. William
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Theory and Modeling in Nanoscience

Report of the May 10–11, 2002, Workshop
Conducted by the Basic Energy Sciences
and Advanced Scientific Computing
Advisory Committees to the Office of Science,
Department of Energy

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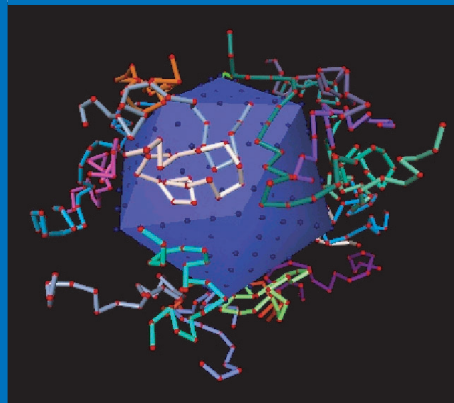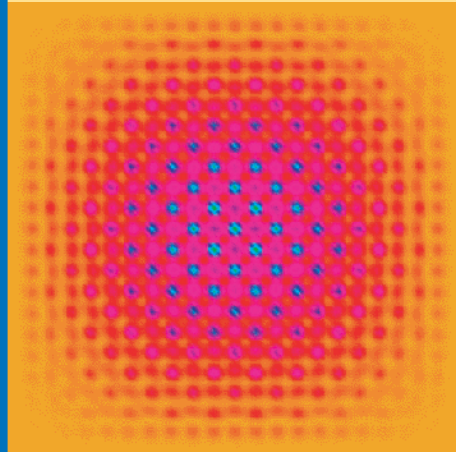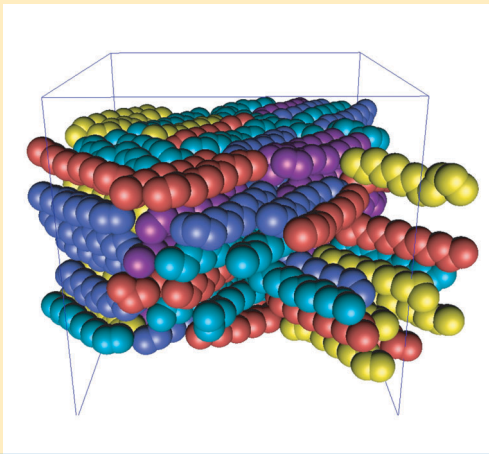
Cover illustrations:

TOP LEFT: Ordered lubricants confined to nanoscale gap (Peter Cummings).

BOTTOM LEFT: Hypothetical spintronic quantum computer (Sankar Das Sarma and Bruce Kane).

TOP RIGHT: Folded spectrum method for free-standing quantum dot (Alex Zunger).

MIDDLE RIGHT: Equilibrium structures of bare and chemically modified gold nanowires (Uzi Landman).

BOTTOM RIGHT: Organic oligomers attracted to the surface of a quantum dot (F. W. Starr and S. C. Glotzer).

Theory and Modeling in Nanoscience

**Report of the May 10–11, 2002, Workshop
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Executive Summary

On May 10 and 11, 2002, a workshop entitled "Theory and Modeling in Nanoscience" was held in San Francisco to identify challenges and opportunities for theory, modeling, and simulation in nanoscience and nanotechnology and to investigate the growing and promising role of applied mathematics and computer science in meeting those challenges. A broad selection of university and national laboratory scientists contributed to the workshop, which included scientific presentations, a panel discussion, breakout sessions, and short white papers.

Revolutionary New Capabilities in Theory, Modeling, and Simulation

During the past 15 years, the fundamental techniques of theory, modeling, and simulation have undergone a revolution that parallels the extraordinary experimental advances on which the new field of nanoscience is based. This period has seen the development of density functional algorithms, quantum Monte Carlo techniques, *ab initio* molecular dynamics, advances in classical Monte Carlo methods and mesoscale methods for soft matter, and fast-multipole and multigrid algorithms. Dramatic new insights have come from the application of these and other new theoretical capabilities. Simultaneously, advances in computing hardware increased computing power by four orders of magnitude. The combination of new theoretical methods together with increased computing power has made it possible to simulate systems with millions of degrees of freedom.

Unmistakable Promise of Theory, Modeling, and Simulation

The application of new and extraordinary experimental tools to nanosystems has created an urgent need for a quantitative understanding of matter at the nanoscale. The absence of quantitative models that describe newly observed phenomena increasingly limits progress in the field. A clear consensus emerged at the workshop that without new, robust tools and models for the quantitative description of structure and dynamics at the nanoscale, the research community would miss important scientific opportunities in nanoscience. The absence of such tools would also seriously inhibit widespread applications in fields of nanotechnology ranging from molecular electronics to biomolecular materials. To realize the unmistakable promise of theory, modeling, and simulation in overcoming fundamental challenges in nanoscience requires new human and computer resources.

Fundamental Challenges and Opportunities

With each fundamental intellectual and computational challenge that must be met in nanoscience comes opportunities for research and discovery utilizing the approaches of theory, modeling, and simulation. In the broad topical areas of (1) nano building blocks (nanotubes, quantum dots, clusters, and nanoparticles), (2) complex nanostructures and nano-interfaces, and (3) the assembly and growth of nanostructures, the workshop identified a large number of theory, modeling, and simulation challenges and opportunities. Among them are:

- to bridge electronic through macroscopic length and time scales

- to determine the essential science of transport mechanisms at the nanoscale

- to devise theoretical and simulation approaches to study nano-interfaces, which dominate nanoscale systems and are necessarily highly complex and heterogeneous

- to simulate with reasonable accuracy the optical properties of nanoscale structures and to model nanoscale opto-electronic devices

- to simulate complex nanostructures involving "soft" biologically or organically based structures and "hard" inorganic ones as well as nano-interfaces between hard and soft matter

- to simulate self-assembly and directed self-assembly

- to devise theoretical and simulation approaches to quantum coherence, decoherence, and spintronics

- to develop self-validating and benchmarking methods

The Role of Applied Mathematics

Since mathematics is the language in which theory is expressed and advanced, developments in applied mathematics are central to the success of theory, modeling, and simulation for nanoscience, and the workshop identified important roles for new applied mathematics in the above-mentioned challenges. Novel applied mathematics is required to formulate new theory and to develop new computational algorithms applicable to complex systems at the nanoscale.

The discussion of applied mathematics at the workshop focused on three areas that are directly relevant to the central challenges of theory, modeling, and simulation in nanoscience: (1) bridging time and length scales, (2) fast algorithms, and (3) optimization and predictability. Each of these broad areas has a recent track record of developments from the applied mathematics community. Recent advances range from fundamental approaches, like mathematical homogenization (whereby reliable coarse-scale results are made possible without detailed knowledge of finer scales), to new numerical algorithms, like the fast-multipole methods that make very large scale molecular dynamics calculations possible. Some of the mathematics of likely interest (perhaps the most important mathematics of interest) is not fully knowable at the present, but it is clear that collaborative efforts between scientists in nanoscience and applied mathematicians can yield significant advances central to a successful national nanoscience initiative.

The Opportunity for a New Investment

The consensus of the workshop is that the country's investment in the national nanoscience initiative will pay greater scientific dividends if it is accelerated by a new investment in theory, modeling, and simulation in nanoscience. Such an investment can stimulate the formation of alliances and teams of experimentalists, theorists, applied mathematicians, and computer and computational scientists to meet the challenge of developing a broad quantitative understanding of structure and dynamics at the nanoscale.

The Department of Energy is uniquely situated to build a successful program in theory, modeling, and simulation in nanoscience. Much of the nation's experimental work in nanoscience is already supported by the Department, and new facilities are being built at the DOE national laboratories. The Department also has an internationally regarded program in applied mathematics, and much of the foundational work on mathematical modeling and computation has emerged from DOE activities. Finally, the Department has unique resources and experience in high performance computing and algorithms. The combination of these areas of expertise makes the Department of Energy a natural home for nanoscience theory, modeling, and simulation.

I.   Introduction

A.   The Purpose of the Workshop

On May 10 and 11, 2002, a workshop entitled "Theory and Modeling in Nanoscience" was held in San Francisco, California, supported by the offices of Basic Energy Science and Advanced Scientific Computing Research of the Department of Energy. The Basic Energy Sciences Advisory Committee and the Advanced Scientific Computing Advisory Committee convened the workshop to identify challenges and opportunities for theory, modeling, and simulation in nanoscience and nanotechnology, and additionally to investigate the growing and promising role of applied mathematics and computer science in meeting those challenges. The workshop agenda is reproduced in Appendix A.

A broad selection of university and national laboratory scientists were invited, and about fifty were able to contribute to the workshop. The participants are listed in Appendix B. There were scientific presentations, a panel discussion, and breakout sessions, together with written contributions in the form of short white papers from those participants from the DOE labs. This report is the result of those contributions and the discussions at the workshop.

This workshop report should be read in the context of other documents that define and support the National Nanotechnology Initiative. Those documents describe a broad range of applications that will benefit the principal missions of the Department of Energy, ranging from new materials and the energy efficiencies they make possible, to improved chemical and biological sensing. Key among those reports is the one from the Office of Basic Energy Sciences entitled "Nanoscale Science, Engineering and Technology Research Directions" (http://www.sc.doe.gov/production/bes/nanoscale.html), which points out the great need for theory and modeling. Other nanoscience documents from the Department of Energy can be found at http://www.er.doe.gov/production/bes/NNI.htm, and a number of reports from other agencies and groups are linked to the Web site of the National Nanotechnology Initiative, http://www.nano.gov/.

B.   Parallel Dramatic Advances in Experiment and Theory

The context of the workshop was apparent at the outset. The rapid rise of the field of nanoscience is due to the appearance over the past 15 years of a collection of new experimental techniques that have made manipulation and construction of objects at the nanoscale possible. Indeed, the field has emerged from those new experimental techniques.

Some of those experimental methods, such as scanning tunneling microscopy, created new capabilities for characterizing nanostructures. The invention of atomic force microscopy produced not only a tool for characterizing objects at the nanoscale but one for manipulating them as well. The array of experimental techniques for controlled fabrication of nanotubes and nanocrystals, together with methods to fabricate quantum dots and wells, produced an entirely new set of elementary nanostructures. Combinatorial chemistry and genetic techniques have opened the door to the synthesis

of new biomolecular materials and the creation of nano-interfaces and nano-interconnects between hard and soft matter. Nanoscience arose from the entire ensemble of these and other new experimental techniques, which have created the building blocks of nanotechnology.

Over the same 15-year period, the fundamental techniques of theory, modeling, and simulation that are relevant to matter at the nanoscale have undergone a revolution that has been no less stunning. The advances of computing hardware over this period are universally familiar, with computing power increasing by four orders of magnitude, as can be seen by comparing the Gordon Bell Prizes in 1988 (1 Gflop/s) and 2001 (11 Tflop/s). But as impressive as the increase in computing power has been, it is only part of the overall advance in theory, modeling, and simulation that has occurred over the same period. This has been the period in which:

- Density functional theory (DFT) transformed theoretical chemistry, surface science, and materials physics and has created a new ability to describe the electronic structure and interatomic forces in molecules with hundreds and sometimes thousands of atoms (Figure 1).

- Molecular dynamics with fast multipole methods for computing long-range interatomic forces have made accurate calculations possible on the dynamics of millions and sometimes billions of atoms.

- Monte Carlo methods for classical simulations have undergone a revolution, with the development of a range of techniques (e.g., parallel tempering, continuum configurational bias, and extended ensembles) that permit extraordinarily fast equilibration of systems with long relaxation times.

- New mesoscale methods (including dissipative particle dynamics and field-theoretic polymer simulation) have been developed for describing systems with long relaxation times and large spatial scales, and are proving useful for the rapid prototyping of nanostructures in multicomponent polymer blends.

- Quantum Monte Carlo methods now promise to provide nearly exact descriptions of the electronic structures of molecules.

- The Car-Parrinello method for *ab initio* molecular dynamics with simultaneous computation of electronic wavefunctions and interatomic forces has opened the way for exploring the dynamics of molecules in condensed media as well as complex interfaces.

The tools of theory have advanced as much as the experimental tools in nanoscience over the past 15 years. It has been a true revolution.

The rise of fast workstations, cluster computing, and new generations of massively parallel computers complete the picture of the transformation in theory, modeling, and simulation over the last decade and a half. Moreover, these hardware (and basic software) tools are continuing on the Moore's Law exponential trajectory of improvement, doubling the computing power available on a single chip every 18 months. Computational Grids are emerging as the next logical extension of cluster and parallel computing.

The first and most basic consensus of the workshop is clear: *Many opportunities for discovery will be missed if the new tools of theory, modeling, and simulation are not fully exploited to confront the challenges of nanoscience. Moreover, new investments by the DOE and other funding agencies will be*

required to exploit and develop these tools for effective application in nanoscience.

This consensus is not merely speculation, but is based on recent experience of the role of theory in the development of nanotechnology. Perhaps no example is more celebrated than the role of calculations based on density functional theory in the development of giant magnetoresistance in magnetic storage systems. The unprecedented speed with which this discovery was exploited in small hard disk drives for computers depended on a detailed picture from theoretical simulations of the electronic structure and electron (spin) transport in these systems. Some details of this remarkable story are given in a sidebar to this report (page 11).

Figure 1. Nanotubes break by first forming a bond rotation 5-7-7-5 defect. An application of density functional theory and multigrid methods by Buongiorno Nardelli, Yakobson, and Bernholc, Phys. Rev. B and Phys. Rev. Letters (1998).

C. *The Central Challenge*

The discussions and presentations of the workshop identified many specific fundamental challenges for theory, modeling, and simulation in nanoscience. However, a central and basic challenge became clear. Because of the rapid advance of experimental investigations in this area, the need for quantitative understanding of matter at the nanoscale is becoming more urgent, and its absence is increasingly a barrier to progress in the field quite generally.

The central broad challenge that emerged from discussions at the workshop can be stated simply: *Within five to ten years, there must be robust tools for quantitative understanding of structure and dynamics at the nanoscale, without which the scientific community will have missed many scientific opportunities as well as a broad range of nanotechnology applications.*

The workshop audience was reminded in the opening presentation that the electronics industry will not risk deploying billions of

Figure 2. Calculated current-voltage curve for a novel memory-switchable resistor with 5μ × 5μ junctions. (Stan Williams, Hewlett-Packard)

devices based on molecular electronics, even when they can be built, unless they are thoroughly understood (Figure 2) and manufacturing processes are made predictable and controllable. The electronics industry must

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have new simulations and models for nanotechnology that are at least as powerful and predictive as the ones in use today for conventional integrated circuits before it can chance marketing molecular electronics devices for a myriad of applications. It can be argued that biological applications of nanotechnology will require the same level of quantitative understanding before they are widely applied.

The fundamental theory and modeling on which industry will build those tools does not exist. While it is the province of industry to provide its own design tools, it is the role of basic science to provide the fundamental underpinnings on which they are based. Those fundamental tools for quantitative understanding are also necessary to the progress of the science itself.

D.   Key Barriers to Progress in Theory and Modeling in Nanoscience

Much of the current mode of theoretical study in nanoscience follows the traditional separation of the practice of experiment from the practice of theory and simulation, both separate from the underpinning applied mathematics and computer science. This is not a new observation, nor does it apply only to theory, modeling, and simulation in nanoscience. Nonetheless, it is a particularly problematic issue for this field.
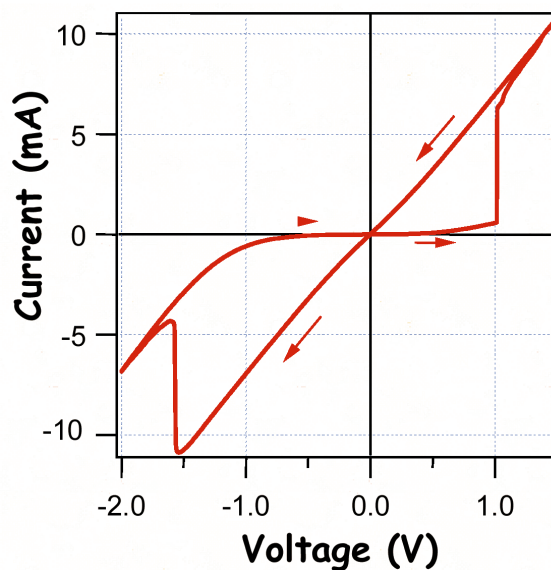
By its very nature, nanoscience involves multiple length and time scales as well as the combination of types of materials and molecules that have been traditionally studied in separate subdisciplines. For theory, modeling, and simulation, this means that fundamental methods that were developed in separate contexts will have to be combined and new ones invented. This is the key reason why an alliance of investigators in nanoscience with those in applied mathematics and computer science will be necessary to the success of theory, modeling, and simulation in nanoscience. *A new investment in theory, modeling and simulation in nanoscience should facilitate the formation of such alliances and teams of theorists, computational scientists, applied mathematicians, and computer scientists*.

A second impediment to progress in theory, modeling, and simulation in nano-

science arises because theoretical efforts in separate disciplines are converging on this intrinsically multidisciplinary field. The specific barrier is the difficulty, given the present funding mechanisms and policies, of undertaking high-risk but potentially high-payoff research, especially if it involves expensive human or computational resources. At this early stage in the evolution of the field, it is frequently not at all clear what techniques from the disparate subdisciplines of condensed matter physics, surface science, materials science and engineering, theoretical chemistry, chemical engineering, and computational biology will be successful in the new context being created every week by novel experiments. The traditional degree of separation of the practice of experiment from the practice of theory further intensifies the challenge.

For these reasons, opportunities will be missed if new funding programs in theory, modeling, and simulation in nanoscience do not aggressively encourage highly speculative and risky research. At least one experimentalist at this workshop complained that high-risk and speculative theoretical and computational efforts are too rare in this field, and that sentiment was echoed by a number of theorists.

E. Consensus Observations

A broad consensus emerged at the workshop on several key observations.

- The role of theory, modeling, and simulation in nanoscience is central to the success of the National Nanotechnology Initiative.

- The time is right to increase federal investment in theory, modeling, and simulation in nanoscience to accelerate scientific and technological discovery.

- While there are many successful theoretical and computational efforts yielding new results today in nanoscience, there remain many fundamental intellectual and computational challenges that must be addressed to achieve the full potential of theory, modeling, and simulation.

- New efforts in applied mathematics, particularly in collaboration with theorists in nanoscience, are likely to play a key role in meeting those fundamental challenges as well as in developing computational algorithms that will become mainstays of computational nanoscience in the future.

F. Opportunity for an Expanded Role for the Department of Energy

The time is ripe for an initiative in theory and modeling of nanoscale phenomena not only because of the magnitude of the potential payoff, but also because the odds of achieving breakthroughs are rapidly improving.

Computational simulation is riding a hardware and software wave that has led to revolutionary advances in many fields represented by both continuous and discrete models. The ASCI and SciDAC initiatives of the DOE have fostered capabilities that make simulations with millions of degrees of freedom on thousands of processors for tens of days possible. National security decisions and other matters of policy affecting the environment and federal investment in unique facilities, such as lasers, accelerators, tokamaks, etc. are increasingly being reliably informed by simulation, as are corporate decisions of similar magnitude, such as where to drill for petroleum, what to look for in new pharmaceuticals, and how to design billion-dollar manufacturing lines. Universi-

ties have begun training a new generation of computational scientists who understand scientific issues that bridge disciplines from physical principles to computer architecture. Advances in extensibility and portability make major investments in reusable software attractive. Attention to validation and verification has repaired credibility gaps for simulation in many areas.

Nanotechnologists reaching toward simulation to provide missing understanding will find simulation technology meeting new challenges with substantial power. However, theorists and modelers working on the interface of nanotechnology and simulation technology are required to make the connections.

The workshop focused considerable attention on the role of applied mathematics in nanoscience. In his presentation, one of the scientists working on molecular dynamics studies of objects and phenomena at the nanoscale expressed the sentiment of the

workshop effectively by saying that the role of applied mathematics should be to "make tractable the problems that are currently impossible." As mathematics is the language in which models are phrased, developments in applied mathematics are central to the success of an initiative in theory and modeling for nanoscience. A key challenge in nanoscience is the range of length and time scales that need to be bridged. It seems likely that fundamentally new mathematics will be needed to meet this challenge.

The diverse phenomena within nanoscience will lead to a plethora of models with widely varying characteristics. For this reason, it is difficult to anticipate all the areas of mathematics which are likely to contribute, and serendipity will undoubtedly play a role. As models and their supporting mathematics mature, new algorithms will be needed to allow for efficient utilization and application of the models. These issues and some significant areas of activity are dis-

cussed in Section 3 of this report. But this discussion is not (and cannot be) fully comprehensive due to the breadth of nanoscience and the unknowable paths that modeling will follow.

The Department of Energy is uniquely situated to build a successful program in theory, modeling, and simulation in nanoscience. Much of the experimental work in nanoscience is already supported by the Department, and new facilities are being built. The Department also has an internationally regarded program in applied mathematics, and much of the foundational work on mathematical modeling and computation has emerged from DOE activities. Finally, the Department has unique resources and experience in high performance computing and algorithms. The conjunction these areas of expertise make the Department of Energy a natural home for nanoscience theory and modeling.

G.   Need for Computational Resources and Readiness to Use Them

The collection of algorithms and computational approaches developed over the last 15 years that form the basis of the revolution in modeling and simulation in areas relevant to nanoscience have made this community intense users of computing at all levels, including the teraflop/s level. This field has produced several Gordon Bell Prize winners for "fastest application code," most recently in the area of magnetic properties of materials. That work, by a team led by Malcolm Stocks at Oak Ridge National Laboratory, is only one example of computing at the terascale in nanoscience.

The workshop did not focus much discussion specifically on the need for computational resources, since that need is ubiquitous and ongoing. The presentations showed computationally intensive research using the

full range of resources, from massively parallel supercomputers to workstation and cluster computing. The sentiment that not enough resources are available to the community, at all levels of computing power, was nearly universally acknowledged. This community is ready for terascale computing, and it needs considerably more resources for workstation and cluster computing.

A significant amount of discussion in the breakout sessions focused on scalable algorithms, meaning algorithms that scale well with particle number or dimension as well as with increasing size of parallel computing hardware. The simulation and modeling community in nanoscience is one of the most computationally sophisticated in all of the natural sciences. That fact was demonstrated in the talks and breakout sessions of

the workshop, and is displayed particularly in the sections of this report devoted to fast algorithms and well characterized nano building blocks.

H. Summary of Specific Challenges and Opportunities

The sections that follow identify a large number of challenges in theory, modeling, and simulation in nanoscience together with opportunities for overcoming those challenges. Because an assembly of 50 scientists and applied mathematicians is too small a number to represent all the active areas of nanoscience and related mathematics, the list compiled at the workshop is necessarily incomplete. However, a summary of even the partial catalog accumulated during the workshop and described briefly in the following sections should make a compelling case for investment in theory, modeling, and simulation in nanoscience. The challenges and opportunities include:

- To determine the essential science of transport mechanisms, including electron transport (fundamental to the functionality of molecular electronics, nanotubes, and nanowires), spin transport (fundamental to the functionality of spintronics-based devices), and molecule transport (fundamental to the functionality of chemical and biological sensors, molecular separations/membranes, and nanofluidics).

- To simulate with reasonable accuracy the optical properties of nanoscale structures and to model nanoscale optoelectronic devices, recognizing that in confined dimensions, optical properties of matter are often dramatically altered from properties in bulk.

- To devise theoretical and simulation approaches to study nano-interfaces, which are necessarily highly complex and heterogeneous in shape and substance, and

are often composed of dissimilar classes of materials.

- To simulate complex nanostructures involving many molecular and atomic species as well as the combination of "soft" biological and/or organic structures and "hard" inorganic ones.

- To devise theoretical and simulation approaches to nano-interfaces between hard and soft matter that will play central roles in biomolecular materials and their applications.

- To address the central challenge of bridging a wide range of length and time scales so that phenomena captured in atomistic simulations can be modeled at the nanoscale and beyond.

- To simulate self-assembly, the key to large-scale production of novel structures, which typically involves many temporal and spatial scales and many more species than the final product.

- To devise theoretical and simulation approaches to quantum coherence and decoherence, including tunneling phenomena, all of which are central issues for using nanotechnology to implement quantum computing.

- To devise theoretical and simulation approaches to spintronics, capable of accurately describing the key phenomena in semiconductor-based "spin valves" and "spin qubits."

- To develop self-validating and benchmarking methodologies for modeling and simulation, in which a coarsergrained description (whether it is

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atomistic molecular dynamics or meso-scale modeling) is always validated against more detailed calculations, since appropriate validating experiments will often be difficult to perform.

For each entry in this list, and the many that can be added to it, there is an established state of the art together with an array of specific technical issues that must be faced by researchers. For all of the challenges listed, there is also an array of fundamental questions to be addressed by researchers in applied mathematics.

This report is a brief synthesis of the effort of approximately 50 experts in the nanosciences, mathematics, and computer science, drawn from universities, industry, and the national laboratories. Following the scientific presentations and a panel discussion on the roles of applied mathematics and computer science, the principal recommendations of the workshop were developed in breakout sessions. Three of them focused on nanoscience directly:

- Well Characterized Nano Building Blocks

- Complex Nanostructures and Interfaces

- Dynamics, Assembly, and Growth of Nanostructures

Three others focused on the role of applied mathematics and computer science:

- Crossing Time and Length Scales

- Fast Algorithms

- Optimization and Predictability

There were, of course, other possible ways to organize the discussions, but the outcome would likely have been the same no matter what the organization of the topics. Nevertheless, the structure of this report reflects this particular selection of categories for the breakout sessions.

Giant Magnetoresistance in Magnetic Storage

The giant magnetoresistance (GMR) effect was discovered in 1988 and within a decade was in wide commercial use in computer hard disks (Figure 3) and magnetic sensors. The technology significantly boosted the amount of information that could be recorded on a magnetic surface (Figure 4). The unprecedented speed of application (less than 10 years from discovery to deployment) resulted largely from advances in theory and modeling that explained the microscopic quantum-mechanical processes responsible for the GMR effect.
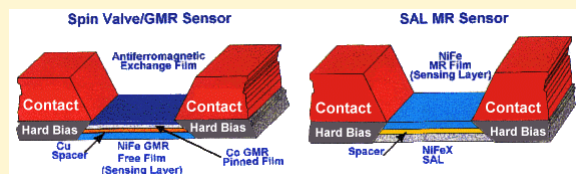

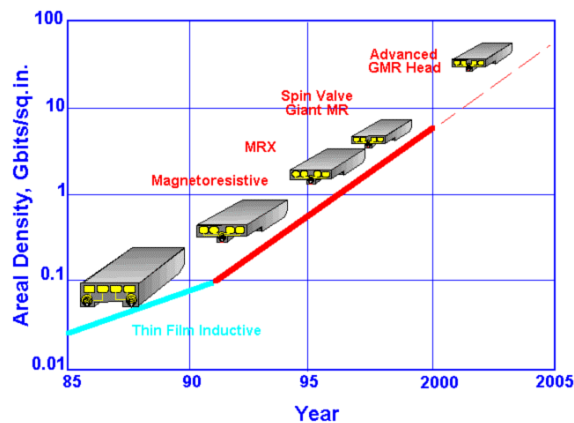**Figure 3. GMR and MR head structures. (IBM)**

Figure 4. Magnetic head evolution. (IBM)

The effect is observed in a pair of ferromagnetic layers separated by a (generally) nonmagnetic spacer. It occurs when the magnetic moment in one ferromagnetic layer is switched from being parallel to the magnetic moment of the other layer to being antiparallel (Figure 5). When a read head senses a magnetic "bit" on the storage medium, it switches the magnetic moment on one layer and measures the resistance, thus sensing the information on the disk.

Soon after the discovery of GMR, the phenomenon was seen to be related to the different "resistances" of electrons having different spins
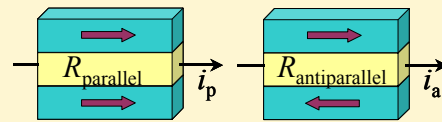

Figure 5. Schematic of GMR indicating change in resistance accompanying magnetization reversal upon sensing an opposing bit. (IBM)

(up or down). In fact, the magnetic moment itself results from an imbalance in the total number of electrons of each spin type. Modern quantum-mechanical computational methods based on density functional theory (for which Walter Kohn was awarded the 1998 Nobel Prize) then provided a detailed picture of the electronic structure and electron (spin) transport in these systems. Indeed, some unexpected effects, such as spin-dependent channeling, were first predicted on the basis of first-principles calculations and were only later observed experimentally.

In fact, GMR is only one aspect of the rich physics associated with the magnetic multilayers now used in read heads. Equally important are oscillatory exchange coupling (which is used to engineer the size of the magnetic field required to switch the device) and exchange bias (which is used to offset the zero of the switching field in order to reduce noise). In exchange coupling, a detailed understanding of the mechanisms responsible has been obtained on the basis of first-principles theory. Less is known about exchange bias, but significant progress has been made on the basis of simplified models and with first-principles calculations of the magnetic structure at the interface between a ferromagnet and an antiferromagnet.

Impressive as these advances in theory and modeling have been, their application to nanomagnetism has only just begun. New synthesis techniques have been discovered for magnetic nanowires, nanoparticles, and molecular magnets; magnetic semiconductors with high Curie temperatures have been fabricated; and spin-polarized currents have been found to drive magnetic-domain walls. When understood through theory and modeling, these findings also are likely to lead to technological advances and commercial applications.

II.   Theory, Modeling, and Simulation in Nanoscience

The challenges presented by nanoscience and nanotechnology are not simply restricted to the description of nanoscale systems and objects themselves, but extend to their design, synthesis, interaction with the macroscopic world, and ultimately large-scale production. Production is particularly important if the technology is to become useful in society. Furthermore, the experience of the participants in the workshop from the engineering community strongly suggests that a commercial enterprise will not commit to large-scale manufacture of a product unless it can understand the material to be manufactured and can control the process to make products within well-defined tolerance limits.

For macroscopic systems—such as the products made daily by the chemical, materials, and pharmaceutical industries—that knowledge is often largely or exclusively empirical, founded on an experimental characterization over the ranges of state conditions encountered in a manufacturing process, since macroscopic systems are intrinsically reproducible. Increasingly, however, this characterization is based on molecular modeling, including all of the tools relevant to modeling nanoscale systems, such as electronic structure, molecular simulation, and mesoscale modeling methods. Indeed, the report *Technology Vision 2020: The U.S. Chemical Industry*[1] has identified molecular modeling as one of the key technologies that the chemical industry needs to revolutionize its ability to design and optimize chemicals and the processes to manufacture them.

Molecular modeling already plays a major role in the chemical, materials, and pharmaceutical industries in the design of new products, the design and optimization of manufacturing processes, and the trouble-shooting of existing processes. However, the exquisite dependence on details of molecular composition and structures at the nanoscale means that attempting to understand nanoscale systems and control the processes to produce them based solely on experimental characterization is out of the question.

The field of nanoscience is quite broad and encompasses a wide range of yet-to-be-understood phenomena and structures. It is difficult to define nanoscience precisely, but a definition consistent with the National Nanotechnology Initiative is:

> *The study of structures, dynamics, and properties of systems in which one or more of the spatial dimensions is nanoscopic (1–100 nm), thus resulting in dynamics and properties that are distinctly different (often in extraordinary and unexpected ways that can be favorably exploited) from both small-molecule systems and systems macroscopic in all dimensions.*

Rational fabrication and integration of nanoscale materials and devices offers the promise of revolutionizing science and technology, provided that principles underlying their unique dynamics and properties can be discovered, understood, and fully exploited. However, functional nanoscale structures often involve quite dissimilar materials (for example, organic or biological in contact with inorganic), are frequently difficult to characterize experimentally, and must ultimately be assembled, controlled, and utilized by manipulating quantities (e.g., temperature, pressure, stress) at the macroscale.

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[1] American Chemical Society et al., 1996, http://membership.acs.org/i/iec/docs/chemvision2020.pdf.

This combination of features puts unprecedented demands on theory, modeling, and simulation: for example, due to nanoscale dimensionality, quantum effects are often important, and the usual theories valid either for bulk systems or for small molecules break down. Qualitatively new theories are needed for this state of matter intermediate between the atomic scale and a large-enough scale for collective behavior to take over, as well as methods for connecting the nanoscale with finer and coarser scales.

Within the context of this definition of nanoscience and the need for significant advances in our understanding of the nanoscale, three breakout sessions on theory, modeling, and simulation in nanoscale science considered three broad classes of nanosystems:

- The first class consisted of nano building blocks (such as nanotubes, quantum dots, clusters, and nanoparticles), some of which can be synthesized quite reproducibly and well characterized experimentally.

- The second class consisted of complex nanostructures and nano-interfaces,[2] reflecting the importance of nano-interfaces in a wide variety of nanoscale systems. This session was specifically asked to consider steady-state properties of complex nanostructures and nano-interfaces.

- The focus of the third session was dynamics, assembly, and growth of nanostructures, and so was concerned with the dynamical aspects of complex nanostructures. Hence, transport properties (such as electron and spin transport and molecular diffusion) of complex nanostructures, as well as the dynamic processes leading to their creation, particularly self-assembly, were central to this session.

The reports presented below are summaries drafted by the session chairs. Given the ostensibly different charges presented to each group, there is remarkable unanimity in their conclusions with respect to the outstanding theory, modeling, and simulation needs and opportunities. Among the common themes are:

- the need for fundamental theory of dynamical electron structure and transport

- algorithmic advances and conceptual understanding leading to efficient electronic structure calculations of large numbers of atoms

- new theoretical treatments to describe nano-interfaces and assembly of nanostructures

- fundamentally sound methods for bridging length and time scales and their integration into seamless modeling environments

- commitment to an infrastructure for community-based open-source codes.

These common themes have been incorporated into the recommendations and challenges outlined in Section I of this report.

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[2] We can distinguish between interfaces in general and nano-interfaces as follows: Typically, an interface separates two bulk phases; we define, consistent with the above definition, a nano-interface as one in which the extent of one or more of the phases being separated by the interfaces is nanoscopic. In nano-interfaces we include nano-interconnects, which join two or more structures at the nanoscale. For nano-interfaces, traditional surface science is generally not applicable.

A.   Nano Building Blocks

Well characterized building blocks for nanoscience need to be created and quantitatively understood for a number of crucial reasons. A key aspect of construction, whether at the macroscopic or microscopic scale, is the notion of a "building block." Office buildings are often made from bricks and girders, molecular components from atoms. Just as knowledge of the atom allows us to make and manipulate molecular species, knowledge of bricks and mortar allows us to construct high-rise buildings.

Well-characterized nano building blocks will be the centerpiece of new functional nanomechanical, nanoelectronic, and nanomagnetic devices. A quantitative understanding of the transport, dynamics, electronic, magnetic, thermodynamic, and mechanical properties is crucial to building at the nanoscale. As an example, current theoretical approaches to nanoelectronics often cannot accurately predict I-V curves for the simplest molecular systems. Without a coherent and accurate theoretical description of this most fundamental aspect of devices, progress in this area will necessarily be limited.

Theoretical studies for these systems will necessarily have to be based on approximate methods; the systems are simply too large and too complex to be handled purely by *ab initio* methods. Even if the problems are tractable, we will make much greater progress with well-validated approximate models. Thus a variety of benchmarking methodologies and validation testing are needed to establish the accuracy and effectiveness of new models and approximate theory.

At the nanoscale, what are the nano building blocks that would play an analogous role to macro building blocks? Several units come to mind which range from clusters less than 1 nm in size to nanoparticles, such as aerosols and aerogels much larger than 100 nm. However, we believe the best-characterized and most appropriate building blocks are:

- clusters and molecular nanostructures

- nanotubes and related systems

- quantum wells, wires, films and dots.

These blocks are well defined by experiment and frequently tractable using contemporary theory. Moreover, they have been demonstrated to hold promise in existing technology. We believe that building blocks would have an immediate impact in the five areas described below.

Transport in Nanostructures: Electronic Devices

Any electronic device needs to control and manipulate current. At the nanoscale, new phenomena come into play. Classical descriptions become inoperable compared to quantum phenomena such as tunneling, fluctuations, confined dimensionality, and the discreteness of the electric charge. At these dimensions, the system will be dominated entirely by surfaces. This presents further complications for electronic materials, which often undergo strong and complex reconstructions. Defining accurate structure at the interface will be difficult. However, without an accurate surface structure, it will be even more difficult to compute any transport properties. Moreover, current-induced changes within the structure may occur. This situation will require the complexity, but not intractable approach, of a careful self-consistent solution in a non-equilibrium environment.

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Optical Properties on the Nanoscale: Optoelectronic Devices

At confined dimensions, optical properties of matter are often dramatically altered. For example, silicon is the dominant electronic material, but it has poor optical properties for optoelectronic devices such as solar cells or lasers. However, at small dimensions the properties of silicon can be dramatically altered; the band gap in silicon can be blue-shifted from the infrared to the optical region. One of the first manifestations of this effect occurs in porous silicon, which exhibits remarkable room temperature luminescence. To properly capitalize on such phenomena, a deeper quantitative understanding of the optical properties of matter will be required.

Optical excitations are especially challenging because most commonly used methods for structural energies, such as density functional theory, are not well suited for excited state properties. The problem is exacerbated for nanoscale systems, where the many-body effects are enhanced by the physical confinement of the excitation.

Coherence/Decoherence Tunneling: Quantum Computing

While silicon dominates electronic devices, there is a fundamental limit to this technology. As the size of a transistor is reduced each year, eventually one will approach the new size regimes where traditional electronics must be viewed in different terms. The approach to this regime will happen in the not too distant future. Current lithographic techniques can create semiconductor chips with characteristic lengths of only a few hundred nanometers; quantum effects are likely to dominate at tens of nanometers. At this latter length scale, transistor technology will be solidly in the regime of quantum phenomena.

Components will eventually function solely through quantum effects, for which we will need the capability to accurately simulate or predict in order to control. It has been suggested that new computers can be devised at nanoscales that will function by using the creation of coherent quantum states. By creating and maintaining such coherent states, it should be possible to store enormous amounts of information with unprecedented security. In order to construct such new quantum computers, new fabrication methods will need to be found and guided by theory. Current technology is far removed from the routine generation of quantum logic gates and quantum networks. Without a better quantitative description of the science at the nanoscale, the emergence of quantum computing will not happen.

Soft/Hard Matter Interfaces: Biosensors

Nanotechnology has the potential to make great advances in medical technology and biosecurity. The technologies for controlling and processing materials for semiconductor devices can be used to make types of nano-interfaces. Nanostructure synthesis and joining of biological or soft matter with traditional semiconductors like silicon would offer new pathways to the construction of novel devices. For example, one could gain the ability to develop new types of sensors directly integrated into current device technology. A number of clear applications such as biosensors for national security, drug delivery and monitoring, and disease detection are plausible.

However, numerous complex and difficult issues must be addressed. These include quantitative descriptions of nanofluids, reaction kinetics, and a selectivity to particular biological agents. Perhaps no systems are as complex at the nanoscale as these, because they require the understanding of the details of interactions between quite diverse systems, i.e., soft, biological materials inter-

faced with semiconductors or other in organic solids.

Spintronics: Information Technology

Spintronics centers on controlling the spin of electrons in addition to controlling other transport properties. For example, dilute magnetic semiconductors have attracted considerable attention, because they hold the promise of using electron spin, in addition to charge, for creating a new class of spintronic semiconductor devices with unprecedented functionality. Suggested applications include spin field effect transistors, which could allow for software reprogramming of the microprocessor hardware during run time; semiconductor-based spin valves, which would result in high-density, non-volatile semiconductor memory chips; and even spin qubits, to be used as the basic building block for quantum computing.

At the nanoscale, spintronic devices offer special challenges owing to the enhanced exchange terms arising from quantum confinement. Few quantitative studies exist on magnetic properties of quantum dots and the evolution of ferromagnetic properties with size.

Implications for Theory and Modeling

These five areas of research offer great promise but will require new theories (approximate models) and computationally intensive studies. Current algorithms must be made more efficient and sometimes more accurate, or new algorithms must emerge in order to address the challenges highlighted above. For example, *ab initio* methods can address systems with hundreds of atoms, and empirical methods can handle tens of thousands of atoms. To fully exploit these approaches, algorithms need to be made scalable and take full advantage of current computer technology, i.e., highly parallel environments. Fully *ab initio* methods are

especially useful in providing benchmarks for these building blocks, where experimental data is lacking or is unreliable due to unknown variability or uncontrolled critical function characteristics. Embedding methods whereby one can take the best features of a particular approach will be crucial.

Algorithms for particular aspects of nanoscale systems need to be developed. For example, *ab initio* methods for optical properties such as GW and Bethe-Salpeter methods are known to be accurate and robust, yet these systems are limited in their applicability to rather small systems, e.g., to systems of less than 50 atoms. The essential physical features of these methods need to be adapted to address much larger systems.

Nonequilibrium and quantum dynamics present special problems. These systems often lack clear prescriptions for obtaining reliable results. Some recent attempts to match experiments on electron transport through molecular systems often get the correct transport trends, but not quantitative numbers. A number of entanglements exist: Is the experimental data reliable owing to problems with contacts? Or is there some fundamental component missing from our understanding? New experiments will need to be designed to ensure reproducibility and the validity of the measurements. New theories will need to be constructed and cross-checked by resorting to the fundamental building blocks.

A chronic and continuing challenge arising in most nanosystems concerns a lack of understanding of structural relationships. Often nanosystems have numerous degrees of freedom that are not conducive to simple structural minimizations. Simulated annealing, genetic algorithms, and related techniques can be used for some systems, but in general it will require new optimization techniques in order to obtain a quantitative

description of the structure of nanoscale systems.

While great strides have been made in simulation methods, a number of fundamental issues remain. The diversity of time and length scales remains a great challenge at the nanoscale. Rare event simulation methods will be essential in describing a number of phenomena such as crystal growth, surface morphologies, and diffusion. Of course, the transcription of quantum force fields to classical interatomic potentials is a difficult task. Intrinsic quantum at-

tributes like hybridization and charge transfer remain a challenge to incorporate into classical descriptions. At best, current classical-like descriptions can be used for targeted applications.

In order to facilitate dissemination of new algorithms and numerical methods, open-source software should be encouraged and made widely available. Toolkits and efficient codes should be offered without restrictions for distribution to as wide an audience as possible.

B.   Complex Nanostructures and Interfaces

Central to nanoscience is the assembly and manipulation of fundamental building blocks on the nanoscale to create functional structures, materials, and devices (Figure 6). Unlike traditional materials, nanostructured materials that will enable the nanotechnology revolution will be highly complex and heterogeneous in shape and substance, composed of dissimilar materials, and dominated by nano-interfaces (Figure 7). These characteristics require that new theoretical approaches and computational tools be developed to provide a fundamental understanding of complexity at the nanoscale, the physics and chemistry of nano-interfaces, and how interfaces and complexity at the nanoscale control properties and behavior at larger scales.

Nano-interfaces and complexity at the nanoscale play a central role in nearly all aspects of nanoscience and nanotechnology. For example, in molecular electronic devices, DNA and other biomolecules such as proteins are being explored both as assemblers of molecular components like carbon nanotubes and semiconductor quantum dots (Figure 8), and as active components themselves (Figures 9 and 10). In such devices, soft biomolecular matter and hard matter

will come together at complex nano-interfaces across which transport of charge may occur, or which may provide structural stability. In new spintronic devices, the nano-interfaces between ferromagnetic and antiferromagnetic domains may control the speed of switching needed for faster computers. In polymer nanocomposites, where nanoscopic particles are added to a polymer, interactions at the polymer/particle nano-interface can have profound effects on bulk properties, leading to stronger yet lighter-weight structural and other materials. The manipulation of interfaces in soft matter systems at the nanoscale provides a wealth of opportunities to design functional, nano-structured materials for templating, scaffolds for catalysis or tissue growth, photonic devices, and structural materials (Figure 11).

The high surface-to-volume ratio resulting from the prevalence of nano-interfaces, combined with the complexity of these nano-interfaces and of nanostructures, provides much of the challenge in developing predictive theories in nanoscience. In this respect, materials designed at the nanoscale are distinctly different from traditional bulk materials. One of the largest driving forces behind the need for new theory and simula-
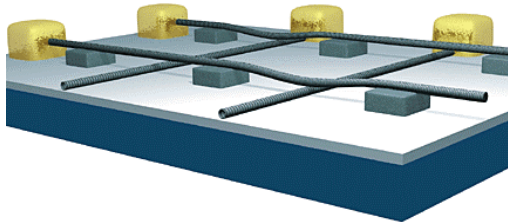
Figure 6. Three-dimensional view of Charles Lieber's concept for a suspended crossbar array shows four nanotube junctions (device elements), with two of them in the "on" (contacting) state and two in the "off" (separated) state. The bottom nanotubes lie on a thin dielectric layer (for example, SiO₂) on top of a conducting layer (for example, highly doped silicon). The top nanotubes are suspended on inorganic or organic supports (gray blocks). Each nanotube is contacted by a metal electrode (yellow blocks). (IBM)
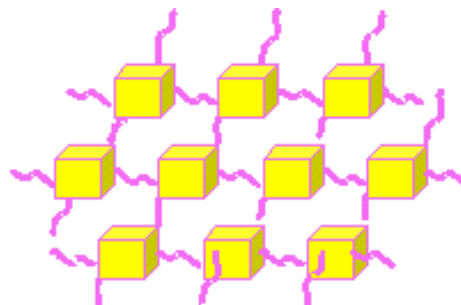
Figure 7. Schematic of an assembled structure of nanoscopic building blocks tethered by organic "linkers." The structure is dominated by interfacial connections. (S. C. Glotzer, 2002)
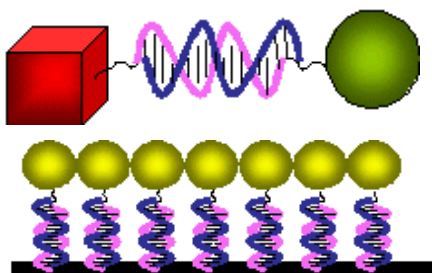
Figure 8. Top: Schematic of nanoscale building blocks tethered by DNA. The "soft/hard" interface in this complex nanostructure controls the transport of information through the structure. Bottom: Schematic of nanoscopic gold nanoparticles assembled onto a surface with DNA. Such guided assembly can be used, e.g., to position the nanoparticles for fabrication into a nanowire.
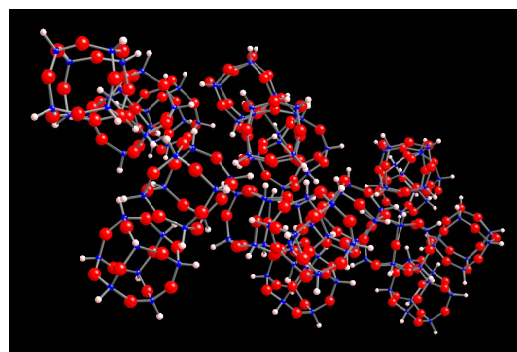
Figure 9. Atomistic simulation of a nanostructure composed of polyhedral oligomeric silsesquioxane cages. (J. Kieffer, 2002)
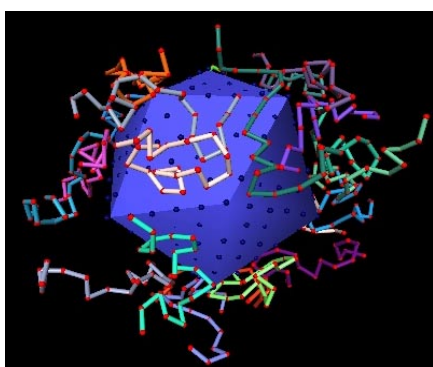
Figure 10. Simulation of organic oligomers attracted to the surface of a nanoscopic quantum dot. (F. W. Starr and S. C. Glotzer, 2001)

Figure 11. Simulation of nanostructured domains in a polymer blend where the interfaces are controlled at the nanoscale. (S. C. Glotzer, 1995)

tion in nanoscience is the fact that there are few experimental techniques capable of directly imaging or probing nano-interfaces. Typically, those techniques having atomic-scale resolution require extensive theory to produce more than qualitative information. Indeed, it is likely that the fundamental interfacial and spatial information required to predict the behavior and performance of nanostructures will come from simulation and theory in many cases.
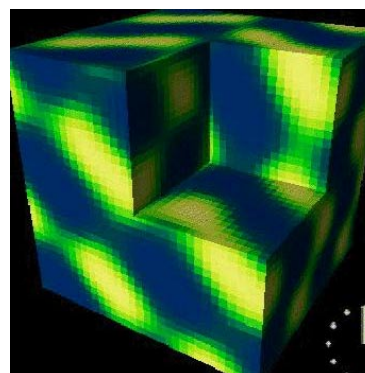
Focused theory and simulation at a wide range of scales is needed to develop a quantitative understanding of nano-interfaces and their consequent influence on properties and emergent behavior of nanoscale materials and devices. Breakthroughs in theory and simulation will also be necessary in order to describe and predict the complexity of nanostructures and the relationship between structural complexity and emergent properties and behavior.

A major challenge for nanoscience in which theory, modeling, and simulation will play a pivotal role lies in the *definition* of the nano-interface, which is nontrivial because the definition may depend upon the properties or behavior of interest. For example, the relevant nano-interface leading to a particular mechanical behavior may be different than that for transport properties, such as electrical transport. Strategies for addressing nano-interfaces will also be needed. With an appropriate definition of nano-interfaces, new approaches can be developed that characterize relevant nano-interfaces in order to construct quantitatively predictive theories that relate the nano-interfaces to interesting nanoscale properties and behavior.

To develop theories and quantitative understanding of nano-interfaces, it will be necessary to obtain information on the organization of matter at nano-interfaces, that is, the arrangement, composition, conforma-tion, and orientation of matter at hard-hard, hard-soft, and soft-soft nano-interfaces. Much of this information may only be accessible by simulation, at least for the foreseeable future. The geometry and topology of nano-interfaces, bonding at nano-interfaces, structure and dynamics at or near a nano-interface, transport across nano-interfaces, confinement at nano-interfaces, deformation of nano-interfaces, activity and reactivity at nano-interfaces, etc. must all be investigated by simulation. Such information is needed before very accurate theories relating all of these to material and device operations and behavior will be fully formulated and validated.

For example, to properly design, optimize and fabricate molecular electronic devices and components, we must quantitatively understand transport across soft/hard contacts, which in turn will likely depend on the arrangement of components on the surface and the complexity of structures that can be assembled in one, two, and three dimensions. Comprehending transport will likely require a quantitative understanding of the nature of nano-interfaces between biological matter and hard matter, between synthetic organic matter and hard matter, and between biological matter and synthetic organic matter. It will also require optimization of structures and tailoring of nano-interfaces, which in turn will require a quantitative understanding of the stability of nano-interfaces.

As we move towards the goal of first-principles-based fully theoretical prediction of the properties of nanoscale systems, there is an urgent need to critically examine the application of bulk thermodynamic and statistical mechanical theories to nanostructures and nano-interfaces, where violations of the second law of thermodynamics have been predicted and observed, and where mechanical behavior is not described by continuum theories.

Theories are required that abstract from behavior at a specific nano-interface to predict the behavior of extended structures or material that is comprised of primarily nano-interfaces. Additionally, when describing nanostructures that are collections of a countable number of fundamental building blocks, one must question whether the concept of temperature has the same meaning as in a bulk system. An important task for theoreticians and simulationists will be to identify what can be brought to nanoscience from traditional methods of investigation and description, and then determine what new theories or approaches must be invoked at the nanoscale.

When materials or devices are dominated by nano-interfaces, new physics is likely to result. Examples of nano-interface-dominated physics include the electric field at surfaces, luminescence of rare-earth orthosilicates, surface-enhanced Raman scattering, and deformation of nanocrystalline materials. Nanocomposites and nano-assemblies are just two examples of nano-structures where nano-interfaces are prevalent and may dominate behavior at larger scales. The high surface-to-volume ratio of complex nanostructures may also lead to novel collective or emergent phenomena, which would require new theory and simulation efforts to address and understand.

Because of the present lack of theories to fully describe nano-interfaces at the nanoscale and the ramifications of nano-interfaces on larger scales, and the lack of experimental data on nano-interfaces, simulations will play a pivotal role in advancing knowledge in this area. *Ab initio* calculations in particular will be required, but the present state of the art limits the number of atoms to numbers too small to obtain meaningful data in many instances. Consequently, breakthroughs in methodologies will be required to push *ab initio* simulations to larger scales. *Ab initio* simulations containing of the order of 10,000 atoms would enable, for example, the study of spin structure at antiferromagnetic-ferromagnetic nano-interfaces.

New or improved computational methods are needed to simulate also the dynamics of nano-interfaces from first principles. Such simulations will be needed to predict, for example, electronic transport across nano-interfaces, as well as to provide validation or benchmarks for coarser-grained theoretical descriptions of nano-interfaces in which electronic structure is not explicitly considered. For molecular dynamics methods and other classical, particle-based methods, new (for many purposes reactive) force fields capable of describing nano-interfaces between dissimilar materials are urgently needed. For some problems, further coarse-graining will be required to capture complexity on all relevant length and time scales, and thus new mesoscale theories and computational methods will be needed. To connect the properties and behavior of complex nanostructures and/or nano-interfaces to properties and behavior at the macroscale, it may be necessary to develop new constitutive laws (and ask if it is even appropriate to talk about constitutive laws in systems where properties and behavior are dominated by nano-interfaces) and then develop and integrate theories at different levels.

C. Dynamics, Assembly, and Growth of Nanostructures

The focus of this section is the time-dependent properties of nanostructures (such as transport properties) and the time-dependent processes used to produce nanostructures and nanostructured materials (such as directed self assembly, nucleation and growth, and vapor deposition methods).

The primary route to manufacturable functional nanoscale systems is universally conceded to be self-assembly, which may be directed or not. Self-assembly of nanostructured materials is scalable to industrial levels of production because it does not require control at the nanoscale (such as, for example, manipulation via AFM tip). There is a wide variety of transport mechanisms relevant to nanoscience, including electron transport (relevant to molecular electronics, nanotubes, and nanowires), spin transport (relevant to spintronics-based devices, see Figure 12), and molecule transport (relevant to chemical and biological sensors, molecular separations/membranes, and nanofluidics).

Examples of nanostructures and nanostructured systems formed by (or capable of formation by) self-assembly include quantum dot arrays, nano- and microelectromechanical systems (NEMS and MEMS), nanoporous adsorbents and catalytic materials (produced by templated self-assembly, see Figure 13), nanocrystals, and biomimetic materials. This list is by no means exhaustive.

The role that mathematics and computer science researchers can play is to apply mathematical and computational tools developed in other contexts to the understanding, control, design, and optimization of nanostructured materials and functional systems based on nanoscale structures. Each of these elements—understanding, control,
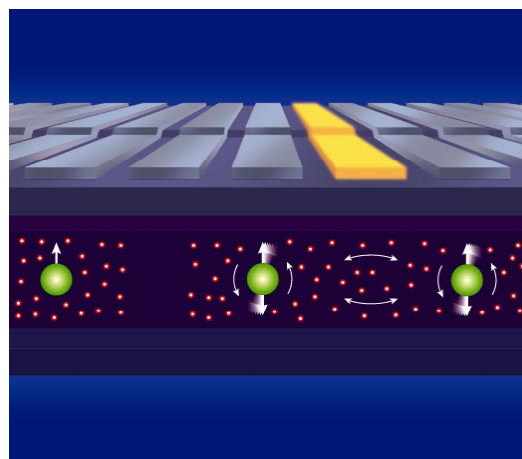
Figure 12. Spintronics devices utilize an electron's spin rather than its charge. Shown here is an artist's depiction of a proposal by Bruce Kane of the University of Maryland for a quantum computer based on the nuclear spin of phosphorus atoms (green), which interact by means of spin-polarized electrons (red). The quantum properties of superposition and entanglement may someday permit quantum computers to perform certain types of computations much more quickly using less power than is possible with conventional charge-based devices. (From S. Das Sarma, "Spintronics," *American Scientist* 89, 516 (2001))
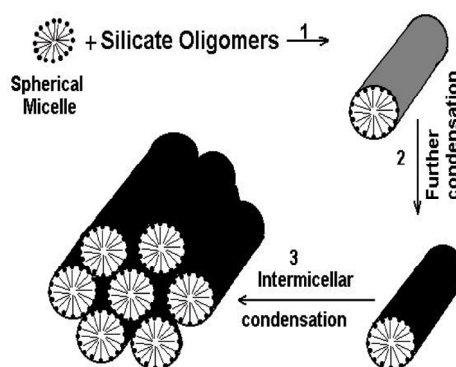
Figure 13. Templated self assembly of nanoporous material—in this case, MCM-41. (From X. S. Zhao, "Synthesis, modification, characterization and application of MCM-41 for VOC Control," Ph.D. thesis, University of Queensland, 1998)

design, and optimization—is essential to the manufacturability of systems composed of nanoscale objects, as noted by Stan Williams of Hewlett-Packard in the opening talk of the workshop.

Among the major challenges facing theory, modeling, and simulation (TMS) in the dynamics, assembly, and growth of nanostructures is simulation of self-assembly (which typically involves many temporal and spatial scales, and many more species than the final product), methods for calculating electron and spin transport properties in nanoscale systems, and the processing and control (particularly to achieve more uniform size distribution) of self-assembly of heterostructures composed of hard materials (such as group IV semiconductors, obtained by combined ion and molecular beam epitaxial growth).

The TMS methods needed to faithfully model the dynamics, assembly, and growth of nanostructures consist of the usual methods we associate with TMS (electronic structure methods, atomistic simulation, mesoscale and macroscale methods) but with additional variations specific to these problems. These variations include:

- Hybrid electronic structure/molecular dynamics methods scalable to very large systems are needed to model combined reaction and structure evolution.

- Coarse graining/time spanning methods are crucial, owing to the large times associated with self-assembly and the large microscopic and mesoscopic domains that exist in many systems.

- Since many self-assembly processes take place in solution, electronic structure methods incorporating solvation are necessary.

- Molecular and nanoscale methods that incorporate phase equilibria and non-

equilibrium multiphase systems (including interfaces) are required.

Participants also saw the necessity of traditional finite element elastic/plastic codes for stress/strain description at the macroscale, since ultimately nanostructured materials must connect with the macroscopic world. It was noted that upscaling (going to coarser length and time scales) typically results in the introduction of stochastic terms to reflect high-frequency motion at smaller scales; thus, more rigorous methods for including stochasm and more effective methods for characterizing and solving stochastic differential equations are required.

New developments in nonequilibrium statistical mechanics, both of classical and quantum systems, are needed. For example, the recent discovery by nonequilibrium molecular dynamics, confirmed by nonlinear response theory, of violations of the second law of thermodynamics for nanoscale systems indicates the importance of new theoretical developments focused on nanoscale systems. Quantifying errors in calculated properties was identified as a significant problem that consists of two parts—characterization of uncertainty due to inaccuracies inherent in the calculation (such as limitations of a force field in molecular dynamics or of a basis set in electronic structure calculations) and performing useful sensitivity analyses.

The important aspect of nanoscale systems that makes them such a TMS challenge is that the characterization of uncertainty will often have to be done in the absence of experimental data, since many of the property measurement experiments one would like to perform on nanoscale systems are impossible in many cases or unlikely to be done in cases where they are possible. Hence, the concept of self-validating TMS methods arises as a significant challenge in

nanoscience. By self-validating TMS methods we mean that a coarser-grained description (whether it be atomistic molecular dynamics or mesoscale modeling, for example) is always validated against more detailed calculations (e.g., electronic structure calculations in the case of atomistic molecular dynamics, and atomistic molecular dynamics in the case of mesoscale modeling).

Various computational issues were identified as important for computational nanoscience. In particular, an open source software development framework for multiscale modeling was identified as a real need. The OCTA project in Japan (http://octa.jp) may point the way towards development of such an open source software development framework, or may indeed be the appropriate framework on which to expand simulation capabilities. Visualization of nanoscale systems is challenging, since the spatiotemporal scales cover such large orders of magnitude. Truly effective parallelization of Monte Carlo was identified as an important issue. Specifically, efficient domain decomposition parallelization of Monte Carlo on very large systems with long range forces was seen as an unsolved problem with ramifications for computational nanoscience. Finally, parallel codes for any TMS method that scales efficiently to tens of thousands of processors were targeted. This is because the highest-performance computers are headed towards this number of processors, yet it is not clear that any scalable codes exist at this point for such large machines.

We identified three grand-challenge-scale problems within self-assembly that would require theoretical, algorithmic, and computational advances, and which might be specifically targeted in any future program in computational nanoscience. These are the simulation of self-assembly of templated nanoporous materials, self-assembly of nanostructures directed by DNA and DNA-like synthetic organic macromolecules, and directed self-assembly of quantum dot arrays and other nanoscale building blocks using physical synthesis and assembly methods. Each of these problems has clear relevance to the DOE mission, creating possible paradigm shifts in low-energy separations, catalysis, sensors, electronic devices, structural and functional hybrid materials, and computing. Each involves significantly different chemical and physical systems, so that the range of TMS methods needed varies with each problem: the first involves self-assembly from solution; the second the use of biologically or bio-inspired directed self-assembly, again in solution; while the third generally does not involve solutions.

In a related example, Figure 14 shows a new computational method for producing random nanoporous materials by mimicking the near-critical spinodal decomposition process used to produce controlled-pore glasses (CPGs). While producing model CPGs which have similar pore distributions and sizes to actual CPGs, the method is not a fully detailed simulation of CPG synthesis. Refinement of such techniques to the point where new nanoporous materials can be discovered and synthesized computationally would be a paradigm-altering step forward in nanostructured materials synthesis.
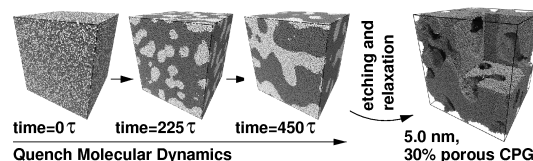
Figure 14. Schematic of controlled-pore glass model preparation. The phase separation process used to prepare real materials is imitated via molecular dynamics simulation, resulting in molecular models with realistic pore networks and structures. (After Gelb et al., Reports On Progress in Physics, Vol. 62, 1999, p. 1573–1659)

III.   The Role of Applied Mathematics and Computer Science in Nanoscience

"I am never content until I have constructed a mechanical model of what I am studying. If I succeed in making one, I understand; otherwise I do not. . . . When you measure what you are speaking about and express it in numbers, you know something about it, but when you cannot express it in numbers your knowledge about it is of a meagre and unsatisfactory kind."
—William Thompson (Lord Kelvin), 1824–1907

Kelvin's mathematical triumphalism is from a pre-quantum-mechanical era and must be discounted as chauvinistic in view of the immense understanding of complex phenomena that can be accumulated *prior* to satisfactory mathematization. Such hard-won understanding of phenomena, in fact, *leads* eventually to their mathematization. Nevertheless, Kelvin's agenda of quantitative understanding remains as a prerequisite for simulation and predictive extrapolation of a system, if not for substantial understanding.

Given the enormous power of computational simulation to transform any science or technology supported by a mathematical model, the mathematization of any new field is a compelling goal. The very complexity and breadth of possibilities in nanoscience and technology cry out for an increasing role for computational simulation today. This section, organized around three themes, indicates some of the directions this increasing role might take: in bridging time and length scales, in fast algorithms, and in optimization and predictability. In each of these rep-

resentative areas, there are instances of "low-hanging fruit," of problems that are reasonably well defined but whose solutions present significant technical challenges, and of phenomena that are not yet at all well defined by Kelvinistic standards.

In a complementary way, mathematics and simulation would be enormously stimulated by the challenges of nanoscale modeling. While some rapidly developing areas of mathematics, such as fast multipole and multigrid algorithms, are ready to apply (in fact, are already being used) in nanoscale modeling, there is much work to do just to get to the frontier of the science in other areas.

It is important to keep in mind that no one can predict where the next mathematical breakthrough of consequence to nanoscience will come from: it could be from computational geometry or from graph theory; it might come from changing the mathematical structure of existing models. Changing the mathematical structure creates a new language, which often can expose an emergent simplicity in a seemingly complex landscape. For instance, no one could have predicted the significance of space-filling curves (a topological fancy) to the optimal layout of data in computer memories.

However, it is predictable that the forward progress of mathematical modeling will carry first one, then another area of nanoscience forward with it—provided that the minds of nanoscientists and mathematicians alike are prepared to experiment, to look for the points of connection, and to recognize opportunities as they arise. The many obvious connections described below

should not overshadow the potential for the emergence of perhaps more important connections between nanoscience and other areas of mathematics in the near future.

Hence, it will be important at this early stage not to define too narrowly what forms of applied mathematics will be important to nanoscience applications. Furthermore, the collaborative infrastructure and cross-fertilization between projects that can be facilitated in a DOE initiative fosters creativity while making recognition of otherwise non-obvious connections and opportunities more likely.

One of the simplest initial objectives of a collaborative dialog between scientists of the nanoscale regime and mathematical modelers is to be sure that the low-hanging fruit is regularly plucked. Mathematical results and software libraries to implement them—many sponsored by the Department of Energy—arrive in a steady stream and readily find application in fields that traditionally engage applied mathematicians, such as engineering mechanics, semiconductor device physics, signal processing, and geophysical modeling. The differential and integral operator equations underlying mathematical physics have been placed on a firm foundation. Issues of existence and uniqueness, convergence and stability of numerical approximation, accuracy and algorithmic complexity, and efficiency and scalability of implementation on the agency's most powerful computers are well understood in many scientific areas.

The variety of simulation tools available is enormous and includes Eulerian-type and Lagrangian-type adaptive methods for the evolution of fields, level sets, and particles. Complex multiscale algorithms have been developed for optimal solution of equilibrium and potential problems. Wonderful tools exist for creating low-dimensional representations of apparently high-information

content phenomena. Stochastic modeling techniques pull averages and higher moments from intrinsically nondeterministic systems. Moreover, the limits of applicability of these tools and techniques are well understood in many cases, permitting their use in a cost-effective way, given a requirement for accuracy. Sensitivity and stability analyses can accompany solutions.

The interaction of scientists and engineers with applied mathematicians leads to continual improvement in scientific and mathematical understanding, computational tools, and the interdisciplinary training of the next generation of each. Due to the novelty of nanoscale phenomena, such interaction with applied mathematicians is often missing. Interactions must be systematically improved in existing and new areas.

At the next level of objectives for a nanoscience-mathematics collaboration lie the problems that are apparently well defined by nanoscientists but for which there are no routinely practical mathematical solutions today. The number of particles that interact, the number of dimensions in which the problem is set, the range of scales to be resolved, the density of local minima in which the solution algorithm may be trapped, the size of the ensemble that needs to be examined for reliable statistics, or some other feature puts the existing model beyond the range of today's analysts and the capacities of today's computers. In these areas, some fundamentally new mathematics may be required.

Finally, there exists a set of problems for which mathematicians must come alongside nanoscientists to help define the models themselves. A critical issue for revolutionary nanotechnologies is that the components and systems are in a size regime about whose fundamental behavior we have little understanding. The systems are too small for easily interpreted direct measurements, but

too large to be described by current first-principles approaches. They exhibit too many fluctuations to be treated monolithically in time and space, but are too few to be described by a statistical ensemble. In many cases, they fall between the regimes of applicability of trusted models. Perhaps existing theories can be stretched. More likely, new theories and hence new mathematical models will be required before robust fundamental understandings will emerge.

Once a theoretical model becomes a trusted companion to experiment, it can multiply the effectiveness of experiments that are difficult to instrument or interpret, take too long to set up, or are simply too expensive to perform. Powerful computational engines of simulation can be called upon to produce, to twist the words of R. W. Hamming, not just *insight*, but *numbers* as well.

Those numbers are the essence of control and predictability.

A. *Bridging Time and Length Scales*

Most phenomena of interest in nanoscience and nanotechnology, like many phenomena throughout science and engineering, demand that multiple scales be represented for meaningful modeling. However, phenomena of interest at the nanoscale may be extreme in this regard because of the lower end of the scale range. Many phenomena of importance have an interaction with their environment at macro space and time scales, but must be modeled in any first-principles sense at atomistic scales.

The time scale required to resolve quantum mechanical oscillations may be as small as a femtosecond (10^{-15} s), whereas protein folding requires microseconds (a range of 10^9), and engineered nanosystems may require still longer simulation periods, e.g., creep of materials relevant to reliability considerations may be measured in years. Counting trials in probabilistic analyses contributes another time-like factor to the computational complexity, although this latter factor comes with welcome, nearly complete algorithmic concurrency, unlike evolutionary time. Meanwhile, spatial scales may span from angstroms to centimeters (a range of 10^8). The cube of this would be characteristic of a three-dimensional continuum-based analysis. Counting atoms or molecules in a single physical ensemble easily leads to the same range; Avogadro's number is about 10^{24}.

Exploiting the concurrency available with multiple independent ensembles multiplies storage requirements. Space-time resource requirements are staggering for many simulation scenarios. Therefore, nanoscience poses unprecedented challenges to the mathematics of multiscale representation and analysis and to the mathematics of synthesizing models that operate at different scales.

Existing techniques may certainly yield some fruit, but we are undoubtedly in need of fundamental breakthroughs in the form of new techniques. Faced with similar types of challenges in macroscopic continuum problems, applied mathematicians have devised a wide variety of adaptive schemes (adaptation in mesh, adaptation in discretization order, adaptation in model fidelity, etc.). Bringing these schemes to a "science" has required decades, and is not complete in the sense of predictable error control for a given investment, e.g., for some hyperbolic problems. Probably the greatest range of scales resolved purely by mesh adaptivity to date is about 10 decades in cosmological gravitation problems. Of course, this range is practical only when the portion of the domain demanding refinement is small compared to

the domain size required to include all of the objects of interest or, what is often the more demanding requirement, to graft on to reliably posed boundary conditions in the far field. Migrating such adaptive schemes into production software (typically through community models, sometimes in commercial analysis packages) lags their scientific development by at least an academic generation and usually more. The challenge for nanoscience is therefore not only great, but also urgent. Applied mathematicians being trained today can acquire a doctoral degree without ever having been exposed to nanoscale phenomena.

The mathematical models and algorithms of nanoscience cover the gamut of continuous and discrete, deterministic and random. Ultimately, all simulations are executed by deterministic programs (at least, to within the limits of race conditions on parallel processors) on discrete data. The most capable computer platforms of 2002 (costing in excess of $100M) execute at most tens of teraflop/s (10^{13} floating point operations per second) and store in fast memory at most tens of terabytes and in disk farms at most hundreds of terabytes. Moreover, these machines do not perform efficiently for models at these capability extremes. A typical extreme problem in computational science today encompasses only billions of discrete degrees of freedom (filling storage larger than this with auxiliary data, including geometry, physical constitutive data, and linear algebra workspaces) and runs for days at tens to hundreds of gigaflop/s. Routine problems encompass only millions of degrees of freedom and run for hours at around 1 gigaflop/s.

The wildest extrapolations of Moore's Law for silicon cannot sate the appetite for cycles and memory in nanoscience modeling. It is clear that a brute force, first-principles approach to nanoscale phenomena will not succeed in bridging scales—or perhaps, given the promise of nanotechnology, we should say that it can only succeed through some "spiral" process, whereby nanotechnology yields superior computers, which yield superior nanoscale models, leading to improved nanotechnology and better computers, and so forth.

We begin with a brief inventory of successful scale-bridging models in applied mathematics today.

Mathematical homogenization (or "upscaling")—whereby reliable coarse-scale results are derived from systems with unresolvable scales, without the need to generate the (out of reach) fine scale results as intermediates—is a success story with extensive theory (dating at least to the 1930s) and applications. The trade-off between accuracy and efficiency is well understood. It is particularly fruitful, for instance, in simulating flows through inhomogeneous porous media or wave propagation through inhomogeneous transmissive media with multiple scatterers. One begins by considering just two scales. Given a 2×2 matrix of interactions: "coarse-coarse," "coarse-fine," "fine-coarse," and "fine-fine," where the first member denotes the scale of an output behavior and the second member the scale of the input that drives the behavior, one can formally solve to eliminate the "fine-fine" block. (If the interactions are all linear, for instance, this can be done with block Gaussian elimination.) This leaves a Schur complement system for the "coarse-coarse" block, where the interaction matrix is the original "coarse-coarse" block and a triple product term with the "fine-fine" inverse in the middle. The effect of the triple-product term can often be reliably approximated, yielding a model for coarse scales that incorporates the dynamics of both scales.

In principle, this process can be repeated recursively. The standard theory succeeds when there is good separation of scales be-

tween the inhomogeneous structure to be "homogenized" and the scales of interest in the solution. When there is significant interaction across length scales (when the "coarse-fine" and the "fine-coarse" interactions are large), the computation of effective properties at the coarse scales becomes more difficult. Work continues on such concepts as "multilevel renormalization," and homogenization should certainly be studied for fruitful approaches to nanoscale science.

Another successful and now quotidian means of bridging scales in modeling is the concept of "space sharing" of different models, with different resolution requirements. For instance, crack propagation in a solid can be effectively modeled with adaptive finite elements for linear elasticity in the mid and far fields, grafted onto molecular dynamics models at and immediately surrounding the crack tip and the newly exposed surface. As another example, the Richtmyer-Meshkov instability in fluid mechanics has been studied with a conventional computational fluid dynamics analysis for Navier-Stokes in the mid and far fields and direct simulation Monte Carlo on "lumped" fluid particles at the mixing interface. Atmospheric reentry problems have similarly been successfully modeled with sparse continuum models away from the body and Boltzmann models in the boundary layer at the body surface.

Very significant scientific and engineering progress can be made with such models on today's high-end computers with such two-physics scale bridging. Less dramatically, one can also cite a multitude of successful dual-continuum model approaches with different asymptotic forms of the governing equations in different regimes, e.g., classical boundary layer theory, in which viscosity terms are computed only in regions of steep gradients. In all of these examples, it is important that the dual models have a regime of overlap, where their solu-

tions can be simultaneously represented and joined. In this sense, these approaches are somewhat complementary to that of homogenization. An even less dramatic example, still, is the scenario in which the physical model remains the same throughout the domain, but the mathematical representation of it changes for computational efficiency. The classic example of this is a combination of finite elements to resolve the geometry around a complex object (e.g., scatterer) in a differential equation approach and boundary elements to resolve the far field in an integral equation approach. These space-sharing approaches sometimes (depending upon the difficulty of the application) lack the mathematical rigor of adaptive approaches within a single physical and mathematical formulation, but with care to monitor and enforce conservation properties at the interface, they are practical and possibly to be emulated in nanoscale phenomena.

A somewhat more trivial instance of bridging scales is what we might call "time sharing" of models, in analogy to "space sharing." In such cases, different dynamically relevant physics "turns on" at different points in the simulation, so that it is acceptable to step over fast scales during one period, but necessary to resolve them in another. Modeling ignition in a combustion system provides one industrially important example. Cosmology enters again here with vastly different physics relevant in the very early universe (e.g., first three minutes) than in the long-term evolution.

It is conceivable, with good software engineering, to marry the concepts of space sharing and time sharing of multiple physics models in a large and complex system, so that each region of space-time adaptively chooses the most cost-effective model and advancement algorithm. One concept in the continuum modeling of multirate systems whose applicability to nanoscale multirate systems should be explored is that of im-

plicit integration methods to "step over" fast time scales that are not dynamically relevant to the result, even though they are timestep limiting, due to stability considerations, in explicit integration. Examples abound, including acoustic waves in aerodynamics, gravity waves in climate modeling, Alfven waves in magnetohydrodynamics, and fast reactions between intermediate species in detailed kinetics of combustion.

In these and many other cases, acceptably accurate results on slower (dynamically relevant) time scales can be achieved by making some equilibrium assumption about the fine dynamics in order to filter out the fast scales, enabling the integration to take place in coarse timesteps. The price for this transformation of the equations to be integrated is usually an implicit solve, where a mathematical balance is enforced on each timestep. The algorithmic cost of an implicit integrator is much higher per step than that of an explicit integrator, but one can take many fewer steps to get to a desired point in simulation time. Whether this trade-off is a winning one for the overall simulation depends upon the cost of the implicit solve. As a rule of thumb in large-scale partial differential equations (PDEs), if the fast scales are two orders of magnitude or more faster than the dynamical scales of interest, implicit methods usually pay off. If the ratio of scales is 10 or less, they likely do not. It may be that many ranges of time scales required by nanoscientists (e.g., integration intervals lasting billions of quantum mechanical oscillation periods) can be condensed by implicit algorithmics without loss of any relevant physics, following some statistical or equilibrium assumptions.

Retreating further into simple, but often adequate, scale-bridging scenarios, there are many examples of one-way cascades of models, whereby a fine-scale problem is solved off-line to develop constitutive laws or material properties for a coarser-scale problem.

It is increasingly practical, for instance, to simulate properties of novel alloys or explosives—materials with simple structure compared to fiber composites or nanotube networks—and then to employ these properties as proxies for experimental data in continuum simulations. Sometimes the properties are quite *ad hoc*, for instance, eddy models in turbulence, to account for mixing at larger scales than molecular by an enhancement of the molecular viscosity in turbulent regimes. Of course, the turbulence simulation community, reflecting a time-honored specialty with a modeling history far too complex to summarize in the scope of this section, copes with multiple scales through as many different means as there are modeling groups. Large-eddy simulation, a fairly rigorous approximation, is emerging as very popular, and its practitioners may have fresh ideas when faced with nanoscale phenomena. Source terms in turbulent reacting flows attempt to account for additional reaction intensity along fractally wrinkled flame fronts by parameterizing smoother, resolvable flame front area. Both deterministic and probabilistic tools have proved fruitful in tuning to experiments and linking up with the fundamental conservation principles. This is a very application-specific technology, as will be, undoubtedly, many of those appropriate for bridging nanoscales.

Although it is not a modeling technique but a solution technique, one should also mention multigrid as a paradigm. Multigrid has already found success in molecular dynamics and in electronic structure algorithms and is likely to be embraced across a wider range of nanoscale applications. Multigrid has the beautiful property of conquering numerical ill conditioning (essentially caused by the presence of too many scales in view of the numerics all at once)

by treating each scale on its own terms. This improves linear conditioning and nonlinear robustness of the solution method, and often leads to physical insight as well. For instance, problems in geometric design are often best approached by resolving large-wavelength geometric parameters on coarse grids, and small-wavelength geometric parameters on finer ones. Multigrid depends on a (usually purely mathematical) decomposition into ranges of scales, and on the ability to construct intergrid transfer operators and coarsened versions of the fundamental fine-scale operator on coarser scales.

Another mathematical paradigm that may prove to have a resonance with the nanoscale community is the method known variously as proper orthogonal decomposition or principal component analysis. In both linear and nonlinear problems, it is often possible to represent the underlying dynamics of interest with a relatively small number of dynamical coefficients representing the weight of complex, system-dependent modes. The challenge is to discover the modes of interest, which are almost never as numerous as the number of characteristic basis functions in the finest grid. The finely resolved setting may be required to determine the principal modes, but once found, production computing can be done in a reduced basis.

Some degree of stochasticity is intrinsic at atomistic scales; hence, a Monte Carlo simulation will yield a fluctuation with statistics that must be carried to larger length scales. The numerical analysis of stochastic partial differential equations could enter here, the goal being a coarse solution with the right statistics. The theory of large deviation for stochastic PDEs allows the designing of sampling methods for so-called "rare events." The long time integrations that are burdensome to the accuracy of deterministic models may actually be a benefit to the accuracy of statistical models.

It is axiomatic to a mathematician that there are better ways than brute-force propagation of fine scales to attack a multiscale problem. A mathematician would argue that anyone requiring a billion degrees of freedom for a week of wall clock execution time is running the wrong algorithm. Indeed, the nanoscale community must learn to compute "smarter" and not just "harder." Part of this will come from optimal algorithmics (e.g., linear scaling in DFT), but a larger and yet unknown portion of it will come from intrinsically multiscale models, created collaboratively with mathematical scientists. It is easy to construct scenarios in which such modeling is attractive, and probably necessary. We mention here an example that appears to demand vast bridging of scales for unified scientific understanding, the operation of a biosensor to detect trace quantities of a biologically active molecule.

- At the smallest scale, a molecule can be sensed by the luminescence characteristic of an optical energy gap. Quantum Monte Carlo could be used for a high-accuracy comparison with experiment.

- At the next scale, where one needs to gain a quantitative understanding of surface structure and interface bonding, DFT and first-principles molecular dynamics could be employed.

- At the next higher scale, one would need to gain a quantitative understanding of the atomistic coupling of the molecule to be detected with the solvent that transports it to the sensor. This modeling could be modeled with classical molecular dynamics at finite temperature and classical Monte Carlo.

- Finally, at the macroscale, the solvent itself could be characterized with a continuous model, based on finite elements.

Each phenomenon depends upon the phenomenon at the scale above and the scale

below, which means that there is two-way coupling between mathematically different models at different scales.

The Holy Grail of nanoscale modeling is a seamless bridging of scales with tight bi-directional coupling between every scale-adjacent pair of a sequence of models, from nano through micro and meso to macro. This bridging necessarily must involve scale-appropriate algorithmics at each scale, with well-understood error dependence on the computational resolution in each model, and with error control for information transfer between scales. For optimality, algorithms should be prudent in their use of fine-scale information—using the coarsest scale suitable for obtaining the requisite error in each result.

B. Fast Algorithms

Due to the inherent complexity of nano-structures, theoretical models will generally need to be instantiated as computational simulations. Many such simulations will involve intensive computation, requiring sophisticated algorithms and software running on state-of-the-art parallel computers. Algorithmic advances can be a critical enabling component for progress in nanoscience modeling. New and more efficient algorithms advance scientific frontiers in several ways:

- Larger simulations can be performed. For example, more atoms or basis functions can be included in a model, increasing fidelity and accuracy. Or dynamical models of the same size can be run for longer physical times. Enabling larger simulations is particularly important in the nanosciences to allow methods at one time or length scale to extend into the range of applicability of other, more coarse-grained methodologies in order to validate or pass information to the coarser-grained methodologies.

- Physical models with more detailed science can be simulated, increasing accuracy and reliability and hence confidence that simulation results represent reality.

- With faster kernel computations, new types of analysis become tractable. For instance, fast linear solvers enable the solution of nonlinear systems. Efficient modeling of forward problems allows for the solution of inverse problems. Hence, faster kernel computations enable a sufficient number of simulations to be performed in order to improve the statistical sampling.

The potential benefits from new capabilities in modeling in the nanosciences require continuous advances in the key algorithms that underlie nanoscale computations. This progress may come about in any number of ways. Many of the key algorithms (fast transforms, eigensolvers, etc.) have applications in other settings, but their impact in the nanosciences alone could justify continued development. Entirely new approaches for some of these problems are difficult to anticipate but could have tremendous impact on not only the feasibility of the computations, but also what will be learned from the computations. For example, wavelets or other new bases might be able to reduce the complexity of some nanoscience calculations and may expose features of the science in new ways. An alternative approach for making progress is to specialize a general algorithm to the precise needs of an application. This approach is the most common way to advance the application of algorithms and often allows for dramatic im-

provements in performance. Recent progress in linear scaling methods for electronic structure provides one such example.

Developments in electronic structure calculations provide a particularly telling illustration of the enabling power of algorithmic advances. First-principles DFT methods for this problem traditionally scale as the cube of the system size, N. This rapid growth with system size limits applications of these powerful techniques to systems containing a few hundred atoms on traditional computers. More efficient cubic algorithms and parallel implementations have allowed larger systems to be simulated, but substantial progress is still limited by the N^3 complexity. Recent algorithmic progress is removing this barrier. For system sizes of up to a thousand electrons, *ab initio* plane wave electronic structure codes using fast Fourier transforms now effectively scale as $N^2 \log N$. Developments using local representations are close to achieving linear scaling for very large systems, at least in some application domains.

A second important example comes from molecular dynamics simulations, where advances in fast multipole methods have reduced the time for computing Coulombic or other long-range forces from N^2 to $N \log N$ or N. Recent innovations in time integration for molecular dynamics are enabling simulations for much longer physical times. Continued developments and advances will be needed to enable the necessary simulations for complex nanoscale dynamics.

These types of advances generate new challenges. The asymptotically more efficient algorithms are generally more complex than their antecedents, and in order to have high fidelity and enough accuracy, some may only be more efficient for very large problems. High-quality implementations are a significant undertaking, and parallel versions will likely require additional algorithmic advances. Modern software practices and open-source parallel libraries can have an enormous impact in easing the development of new functionality and then delivering that new capability to scientists in a timely and efficient manner. More generally, the asymptotic complexity of an algorithm is only one of several aspects relevant to an algorithm's potential impact in nanoscience. Algorithmic researchers must also consider ease of implementation, parallelizability, memory locality, and robustness.

In an area as diverse as nanoscience, an enormous range of computational techniques and algorithms have a role to play, and the workshop was able to touch on only a subset of them. The following areas were identified as particularly important. Many of them are generally applicable for a wide range of applications; however, they will be particularly important to enabling discoveries and quantitative understanding in the nanosciences.

Linear Algebra in Electronic Structure Calculations

Simulation methods based on *ab initio* electronic structure will unquestionably be critical in future investigations of the properties of matter at the nanoscale. The capability of such methods to predict structural and electronic properties without prior empirical knowledge or experimental input makes them very attractive. It is therefore an important priority to accelerate the development and the deployment of fundamental algorithms used in large-scale electronic structure calculations.

Electronic structure calculations based on DFT and extension to first-principles molecular dynamics (FPMD) have traditionally benefited greatly from advances in fast algorithms. An example is the fast Fourier transform, which essentially lies at the root of the efficiency of the plane-wave elec-

tronic structure method, and thus of the feasibility of plane-wave-based FPMD. Depending on the numerical approach taken, the solution of the Kohn-Sham equations is either cast as an eigenvalue problem—in which case the non-linearity due to the Coulomb and exchange-correlation interactions is included as an outer loop—or as a constrained optimization problem—in which case the non-linear Kohn-Sham energy functional is directly minimized with respect to all degrees of freedom describing the electronic wavefunctions, with the constraint that one-particle wavefunctions must remain orthogonal.

Thus the development of efficient eigenvalue solvers on one hand, and of constrained optimization algorithms on the other, directly impacts the efficiency of electronic structure codes. Optimization is discussed elsewhere in this report, but the need for fast eigensolvers is pervasive. Some applications require the k lowest eigenpairs where k is large—perhaps in the thousands. But there is also a need for codes that compute "interior" eigenvalues (eigenvalues inside the spectrum), typically near the Fermi level. Also in time-dependent DFT (TDDFT), a large dense matrix is sometimes diagonalized (in which case all eivenvalues/vectors are computed).

Electronic structure calculations will also likely benefit from all advances that enhance our capability to solve PDEs numerically. In particular, the development of grid-based methods, adaptive mesh refinement approaches, grid partitioning methods, non-linear multigrid solvers, domain decomposition methods, etc. may play an important role in future scalable Kohn-Sham solvers. Specifically, the version of TDDFT that uses ordinary differential equation (ODE) schemes requires the solution of hundreds or thousands (one for each ground state) of systems of ODEs (actually time-dependent Schrödinger equation) which are

coupled by the potential. Potentially enormous gains are possible if specialized methods are developed for solving such systems.

A problem shared by all electronic structure methods is that of efficiently solving the Poisson equation. Algorithms for solving this problem are widespread and well developed, although further progress in the complex geometries relevant to nanoscience applications would be welcome. The community of theorists/modelers involved in nanoscience would benefit from better (and publicly available) software for this problem.

Linear scaling methods and related techniques avoid the eigenvalue problem altogether. Although this allows for a great reduction in asymptotic complexity, current methods can lack accuracy and/or be narrowly applicable. A wealth of algorithms have been proposed and explored, and the opportunities for fruitful collaborative interactions are abundant.

Monte Carlo Techniques

Monte Carlo methods are a class of stochastic optimization techniques that are particularly faithful to the transition probabilities inherent in statistical mechanics or quantum physics. This faithfulness to the underlying physics allows them to be applied to many problems, not just those commonly considered as optimization problems. Improvements in the applicability and (especially) the efficiency of Monte Carlo techniques could have a big impact on several aspects of nanoscience modeling.

Quantum Monte Carlo is currently the most accurate method for electronic structure that can be extended to systems in the nanoscience range. It serves as a benchmark and a source of insight. It is capable of highly accurate numerical solution of the Schrödinger equation. This progress has required resolution of the "fermion sign prob-

lem," a challenge to computational mathematics. That resolution has recently been made in principle, but substantially more development is needed to make it practical.

Kinetic Monte Carlo (KMC) methods are an important class of techniques that treat a range of atomistic and aggregate particle phenomena above that of electronic structure. Essentially they use "Metropolis-Hasting" stochastic dynamics, ascribing physical time intervals to the moves. A fundamental question that needs mathematical attention is the relationship of the time scale to the form of the moves used. If this can be solved, then KMC will be both more reliable and much more widely applicable.

Additional important technical issues connected with KMC include the need for efficient scalably parallel approaches, multiscale methods, and efficient quantitative methods for estimating the probabilities and rates of rare events. In many of the applications, the a priori probability of a rate controlling event is very small; appropriate importance sampling or other techniques need to be developed to make such studies practical.

Monte Carlo methods are also widely used in statistical mechanical studies of nanosystems. All of these variants of Monte Carlo can be made more powerful and useful by incorporating developments in computational geometry and by a theory of optimal moves—that is, by mathematical investigation of the dependence of the spectrum of the Markov transition matrix upon the form of trial moves in dynamics of the Metropolis-Hasting. In addition, since many nanosystems are driven more by entropy than by energy, methods that are entropy or free-energy friendly are badly needed: current methodologies for free energy are usually rather awkward.

Equally important is the promise from a recent development in an O(N) Monte Carlo

method which merits substantial additional development. Other deep technical problems include the need for developments in perturbation methods and techniques for treating relativistic effects.

Finally, continued advances and development in embedding, that is, for using a hierarchy of methods of varying accuracy and computational complexity to treat larger systems, are urgently needed.

Data Exploration and Visualization

Large simulations can generate copious quantities of data. A physical scientist is often interested in exploring these large datasets for interesting features. This process is difficult to automate, since often the most important features are unexpected and they tend to be very application specific. Such explorations are valuable because they generally reveal deficiencies in the simulation itself or unanticipated and interesting aspects of the physics. This kind of data-driven exploring requires rich and easy-to-use environments for visualization and data interactivity. Good tools will hide the complexity of the data management from the user and will respond in real time to user interactivity, even though the data analysis may be occurring on a distant parallel machine.

Although there is considerable research activity in this general area, there will be significant benefit from customization to effectively impact the research of specific nanoscientists. On the other extreme, strategies for deriving maximal information from a limited data environment are needed, since experiments may be difficult or expensive to conduct. This will be critical, for example, in getting a handle on management of uncertainty. In both these contexts, strategies that include experiments for the purpose of supporting the theoretical objective are

needed, in contrast, say, to modeling for the purpose of explaining experiments.

Computational Geometry

Considerable geometric complexity is an inherent aspect of many objects from the atomistic to the nanoscale. Examples include the potential energy isosurfaces of a polymer and the structure of a self-assembled nano-composite. Efficient methodologies for representing and manipulating such geometries can have a significant impact on the overall performance of some computations. Computational geometry is a mature area of algorithmic computer science, but the specific geometries arising in nanoscience would benefit from specialized algorithms.

C.   Optimization and Predictability

In virtually every branch of science, there is a need for calculations to estimate the parameters of a system, to find the extremum of an objective function such as minimum energy or maximum entropy states of a system, and to design and control systems. Since mathematical models are often used to guide experimentation and to predict or adapt to behaviors, there is an equally strong need to understand the errors in these models and to be able to make mathematically sound and precise statements regarding the accuracy and precision of the predictions.

Since all of these needs pervade science, it is natural to ask in what way nanoscience is special. Why, for example, cannot existing techniques be used for the models in nanoscience? The short answer to this question is implicit in the earlier sections of this report: the degree of detail and dynamic complexity including wide-ranging, non-separable scales in both space and time make the formulation and solution of problems in this area fundamentally more challenging than most and arguably all problems successfully tackled to date. The payoff to developing a new set of computational models and tools would be better understanding, prediction, design, optimization, and control of complex nanosystems with confidence in the reliability of the results. Successes in this field would almost certainly have impact on other fields that share the complexity of a large range of complex behavior on non-separable temporal and spatial scales.

Optimization

Many of the problems mentioned above rely on formulating and solving optimization problems. For example, in order to gain a quantitative understanding of nanosystems at a fundamental level, it is essential to have the capability to calculate the ground state of the system. This can be formulated as a minimum energy problem where the solution gives the configuration of the particles of the system at the lowest energy, given an energy functional. Complicated nanosystems of interest can have millions to billions of particles, resulting in huge optimization problems characterized by an equally huge number of local minima with energy levels close to the ground state. There is no hope of solving this problem with brute force methods. To make any headway on such problems requires careful formulation of the objective function and the constraints. For example, progress in the protein-folding problem has been achieved by understanding that certain sequences of amino acids in the unfolded molecule almost always end up in a standard form, e.g., an alpha helix. By inducing the optimization path towards this conformation, better results have been attained. Constraints, based on additional knowledge, can also be formulated to further reduce the search space.

Optimization methods that exploit physical insight could have a dramatic impact on efficiency. Monte Carlo methods are discussed elsewhere in this report, but other stochastic or deterministic optimization methods can also be accelerated by domain knowledge. For example, when trying to simulate a novel material, knowledge about the structural properties and energetics of related materials could be used to preferentially steer the computation towards promising regions of the state space. Such methods are likely to be much more efficient than general optimization strategies, but quite limited in their applicability.

Self-assembly is a central feature of nanoscience. Understanding, predicting, and controlling this process is crucial for the design and manufacture of viable nanosystems. Clearly the subsystems involved in this process are assembling themselves according to some minimum energy principle. Once an understanding of the underlying physics is attained, optimization problems can be formulated to predict the final configurations. Since these systems are also huge and likely to have many local minima, a careful development of the models, the constraints, and the algorithms will also be required here.

Given the necessity to create models that incorporate many scales, it has been suggested that a hierarchy of models be created to span this range, i.e., a connected set of models that capture the quantum effects through the micromorphic effects. In this case, the parameters at a given level should be attainable from the next lowest level using parameter-determining techniques. Optimization formulations for such parameter estimation problems are ubiquitous, but constructing the correct error models and the associated optimization problem is not obvious. Simply using least squares may not be appropriate; more general maximum-likelihood estimators will likely be required.

As alluded to above, the design and control of nanosystems is vital to the eventual manufacture of products. The problem here is to optimize the design to achieve a desired characteristic or to control the process so that a given state is reached. In these cases, the constraints of the optimization problem contain the hierarchy of models describing these systems. In particular, the constraints contain coupled systems of partial differential equations (PDE). There is considerable interest in such problems in other fields, e.g., aerodynamics, and some efficient algorithms have been created, but the number of levels involved in nanoscience is far higher than in other fields.

Progress has been made by understanding that these coupled systems of PDEs do not have to be satisfied at each step of the optimization process, but only need to be satisfied in the limit. This has resulted in an optimized solution in a modest multiple of the time required for one nonlinear solve. Such approaches, however, require that the formulation of the optimization problem incorporate some or all of the state variables in addition to the control variables. Effective optimization methods in these cases must have more control over the PDE software than is typically the case, e.g., gradients and/or adjoints must be computed along with other quantities. Thus, the formulation of the models, the algorithms, and the software with the ultimate goal of design and optimization is crucial.

One can easily envision optimization problems arising in many other aspects of nanoscience. For example, methods for shape optimization for determining the optimal geometry of both tools and products, non-smooth optimization due to derivative discontinuities between the models at adjoining levels, and discrete optimization for particles restricted to discrete locations will be needed. All of the developments in these areas, as with those mentioned above,

should be done collaboratively between optimizers and nanoscientists.

Predictability

Although part of the predictability problem can be addressed using optimization techniques (e.g., maximum likelihood), other statistical methodologies will be necessary to develop a final statement of confidence in the answers. First, however, it is important to recognize all of the possible sources of errors in the process. This starts with the recognition that none of our models is exact. And, since most of the models are based on PDEs, these equations cannot be solved exactly. There will be a combination of discretization errors in the formation of the algebraic systems that the computer sees and roundoff errors in the actual computation. There are errors associated with the physical models and errors related to parameters and initial conditions that define these models. Some of these errors are observational, related to field or laboratory data, and some are numerical or modeling errors, related to simulation data. These two types of errors are actually of a similar form, as laboratory errors are often the result of a simulation needed to interpret the experiment. Furthermore, errors at one scale will be propagated to other scales in ways that will be difficult to track and manage.

It is important to develop methods for determining comprehensive and inclusive bounds on the final errors. Such methods include sampling technologies for developing sensitivities to parameters, analytic techniques for propagation of errors, and probability models for numerical or modeling errors in simulations. The predictions of science are not a simple forward map of an input to an output in a simulation. Parameters and input values needed in the forward simulation are determined as an inverse problem through statistical inference based on measurements of outputs. Thus propagation of errors and uncertainty is mapped both ways, from input to output in direct simulation and from output to input in parameter determination. Statistical inference is the tool that connects the two. This inference can be conveniently formulated in a Bayesian framework, so that the inverse problem acquires a probabilistic expression, and disparate data collected from different sources can then be used to reduce uncertainty and to aid in the predictions. The ultimate goal of such an approach is the determination of error bars or confidence intervals associated with a prediction, based on all sources (data and simulation) on which the prediction is based. A necessary intermediate goal is the association of error bars or other measures of quantified uncertainty with simulations. In analogy to error bars, which indicate the precision of an experimental measurement, error bars should be used to quantify the precision and accuracy of a simulation.

Software

Elsewhere in this report, the issue of software has been raised. The need for well-designed, object-oriented, open-source codes that can be used on a variety of platforms is essential to the overall success of the nanoscale initiative. There are simply too many modules necessary for simulation and analysis than can be developed in one place. Modular optimization and statistical algorithms and software need to be developed in such a framework to provide tools for scientific understanding leading to advanced engineering and analysis tools with guaranteed accuracy for design and manufacture.

Appendix A: Workshop Agenda

Friday, May 10, 2002

	Speaker	Title/Subject
8:00 a.m.	Walt Stevens, DOE, Basic Energy Sciences, and Walt Polansky, DOE, Advanced Scientific Computing Research	"The DOE Perspective on Theory and Modeling in the Nanoscience Initiative"
8:30	C. William McCurdy, LBNL	Goals and Purpose of This Workshop

Session Chair: Bill McCurdy

9:00	Stanley Williams Hewlett-Packard Laboratories	"Big Theory as the Engine of Invention for Nanotechnology: Losing the Born-Oppenheimer Approximation"
9:45	Uzi Landman Georgia Institute of Technology	"Small Is Different: Computational Microscopy of Nanosystems"
10:30	**Break**	

Session Chair: Ellen Stechel

11:00	Steven Louie University of California, Berkeley	"Theory and Computation of Electronic, Transport and Optical Properties on the Nanoscale"
11:45	Dion Vlachos University of Delaware	"Bridging Length and Time Scales in Materials Modeling"
12:30 p.m.	**Lunch – Working, Catered in Conference Room**	

Session Chair: David Keyes

1:45	Phil Colella LBNL	"Computational Mathematics and Computational Science: Challenges, Successes, and Opportunities"
2:30	Jerry Bernholc North Carolina State University	"Quantum Mechanics on the Nanoscale: from Electronic Structure to Virtual Materials"
3:15	**Break**	

Session Chair: Peter Cummings

3:45	Sharon Glotzer University of Michigan	"Computational Nanoscience and Soft Matter"
4:30	Alex Zunger National Renewable Energy Laboratory	"Progress and Challenges in Theoretical Understanding of Semiconductor Quantum Dots"
5:15	Adjourn	
7:00	Conference Dinner – Dinner Speaker: Bernd Hamann, University of California, Davis	"Massive Scientific Data Sets: Issues and Approaches Concerning Their Representation and Exploration"

Saturday, May 11, 2002

	Panel	**Subject**
8:30 a.m.	Paul Boggs, SNL/Livermore Jim Glimm, SUNY Stony Brook Malvin Kalos, LLNL George Papanicolaou, Stanford University Amos Ron, University of Wisconsin-Madison Yousef Saad, University of Minnesota	The Role of Applied Mathematics and Computer Science in the Nanoscience Initiative Panel Moderator: Paul Messina, ANL

Breakout Sessions		*Chair*
10:30	Well Characterized Nano Building Blocks	James Chelikowsky, University of Minnesota
	Complex Nanostructures and Interfaces	Sharon Glotzer, University of Michigan
	Dynamics, Assembly and Growth of Nanostructures	Peter Cummings, University of Tennessee
12:00 p.m.	**Lunch – Working, Catered in Conference Room**	
1:00	**Reports from Breakout Sessions**	

Breakout Sessions		*Chair*
1:30	Crossing Time and Length Scales	George Papanicolaou, Stanford University
	Fast Algorithms	Malvin Kalos, LLNL
	Optimization and Predictability	Paul Boggs, SNL/Livermore
3:00	**Reports from Breakout Sessions**	
3:15	Closing Discussion of the Report of the Workshop	
4:30	Workshop Adjourns	

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