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Dear John and Pat,

We are writing to transmit the preliminary findings and recommendations to Basic Energy Sciences of the BESAC Subcommittee on Theory and Computation. The subcommittee has met twice, once in Washington D. C. on February 22, 2004, immediately prior to the last BESAC meeting, and a second time in Chicago to take testimony and discuss input from the scientific community on April 17 and 18. The ideas expressed in this letter are based on those two meetings and on the written and verbal testimony received from the community at our April meeting and via the subcommittee website, https://besac.nersc.gov/. Already, a broad cross section of the scientific community working in the areas supported by BES has responded to our requests and invitations to testify.

## *The role of theory and computation in the BES portfolio*

The data and testimony collected have made it abundantly clear that a strong partnership of experiment with theory, enhanced and expressed by computation, is already a defining characteristic of the current Basic Energy Sciences portfolio. However, it is our charge to identify new opportunities for that portfolio, as well as the possible changes in strategy, which will enhance and ensure its success over the next five to ten years in meeting the evolving scientific challenges of the BES mission.

At the outset we would like to express the view of the subcommittee that theory and computation should be viewed as a unity, and not as competing parts of the portfolio. By "computation" we do not mean simply raw computing power; we mean the deliberately integrated combination of mathematical modeling and analysis, creation of numerical algorithms, software implementation, and visualization of computed results. We also stress that there is often no clear distinction between theory and the formulation of mathematical models. Our preliminary observations and ideas at this point do not separate theory and computation, although you can expect that our ultimate recommendations will specifically and directly address the computational needs of the BES theory community. The theory enterprise within the fields of science supported by the Office of Basic Energy Sciences of DOE is heterogeneous, and properly so, not only with respect to the scientific problems being addressed, but also with respect to research group size and computational resources required. Innovation flourishes at the single-PI level, providing new theories and insights, the development of new computational methods, and the education and training of future theorists. At the other end of the spectrum are the large, often multidisciplinary groups, investigating problems of great complexity. The solution of such problems requires the development and utilization of large-scale computer codes, demanding leadership-class computer hardware and the implementation of new mathematical methods and algorithms designed to make efficient use of the hardware.

A consensus has emerged in the subcommittee that in order to ensure the highest quality scientific return, the wisest investment on the part of the Office of Basic Energy Sciences is to support the complete spectrum of theory activity, coupled with access to the full range of computational resources appropriate to the particular research being pursued. In each of the areas of new scientific opportunity for theory and computation that we identify, we urge you to "treat the whole patient," and avoid making investment decisions that effectively replace one key part of the theory enterprise with another.

## *New opportunities and challenges for the BES theory enterprise*

As we are sure you realize, the diversity of science interests within the BES portfolio is one of its greatest strengths. Although there are still several important scientific areas from which we have not had testimony, at this time we can provide a partial list of some of the exciting opportunities for theoretical and computational investigation within the range of the BES mission. After vigorous discussion, we have reduced a long list of candidates for new opportunities to just eight, which we list below in no particular order. Two overarching themes have emerged from our discussions that broadly characterize most of these opportunities: ***complexity*** and its complement, ***control***, of physical processes and systems.

**Nanoscale materials and phenomena:** providing early insight and guidance, and ultimately quantitative understanding, for the emerging new science and technology of materials designed, fabricated and operating at nano-meter length scales.

> *Theory and computation are essential partners for the new generation of experiments seeking to measure, control and exploit the structure and properties of nano-materials. The complexity of interactions at the nanoscale requires that the DOE*

*Nanoscience Centers have multidisciplinary teams that include theory and computation for the investigation of nano-magnetism, nano-photonics, nano-mechanics, nano-transport, etc.*

**Biomimetic energy processes:** understanding of photochemical and enzymatic processes leading to improved charge transport mechanisms and ultimately to engineering-level control of biomimetic energy production.

*Hydrogen production by biological and biomimetic processes with sunlight as the driving force and water as the electron source is a promising route to a sustainable, clean energy supply, with the potential of ultimately alleviating the dependence of the United States on foreign suppliers. Large-scale electronic structure calculations and dynamics simulations can play a crucial role in the elucidation of the mechanism of hydrogen production by the molecular machinery of photosynthetic organisms, paving the way for the optimization of hydrogen production by molecular biological modification, and the design of synthetic complexes for biomimetic hydrogen production by artificial photosynthesis.*

**Correlated electrons in solids:** developing theories and new computational methods that will allow quantitative understanding of spectacular phenomena such as high $T_c$ superconductivity, colossal magneto-resistance and exotic magnetic phases.

*Advancing the physics and technological applications of transition metal oxides and other narrow band materials requires the quantitative treatment of strongly correlated electronic states in solids, an outstanding problem for which methods such as Dynamical Mean Field Theory and Quantum Monte Carlo are coming into fruition when coupled with high performance computing.*

**Excited electronic states:** improving and developing new computational approaches to understand the structure and dynamics of electronically excited states in molecules, liquids, and solids, and at interfaces, leading to better control of catalysis and radiation-induced processes.

*Understanding how electronic and nuclear motions are coupled by electronic excitations presents a major computational challenge, particularly for condensed phase systems. A detailed elucidation of these processes will enable the development of new systems for photocatalytic production of hydrogen and design of new materials with a controlled response to ionizing radiation. Furthermore, better understanding of excited electronic states probed via X-ray absorption or scattering is essential for the interpretation of spectra generated by experiments at modern synchrotron facilities.*

**Control of energy, matter, and information flow at the quantum level:** directing the outcome of chemically-reactive processes with shaped laser pulses, and tuning the interaction strengths of few-atom or many-body systems using external fields in quantum gases, are just two exciting examples that demand critical theoretical guidance.

> *A new opportunity in quantum control is to elucidate the connection between Bose and Fermi gases and related topics such as high-$T_c$ superconductivity. Deciding whether a quantum computer is physically realizable, and investigating materials for controlling quantum information, both hinge on basic scientific studies for which the necessary tools reside within the BES portfolio.*

**Ultrafast physics:** optimizing new sources of attosecond light pulses and femtosecond electron pulses, and addressing the new physics accessed by those sources holds the promise of revolutionizing the study and control of electron dynamics.

> *Without new theory, experiments such as time-resolved charge transfer in chemical systems, photoelectron spectra of dissociative states observed during the dissociation, and the newly proposed X-ray Raman spectroscopy will be difficult to design and interpret effectively. For example, understanding time-resolved inner shell electron rearrangements requires the treatment of excited molecular states with core holes whose dissociation dynamics competes with Auger and other electron rearrangement processes.*

**Defects in solids:** developing computational methods for bridging the atomic and mesoscopic length scales to understand and control important physical properties of materials such as strength, transport, fatigue, and magnetic hysteresis.

> *Historically, trial and error has led to the introduction of impurities and defects into materials to strengthen metals, to create better magnets, and to reduce corrosion. Today, a central problem is to understand the collective behavior of defects. For example, line defects (dislocations) interact with each other and with other defects in the material, especially grain boundaries, in a complicated and non-linear fashion. Understanding the collective behavior of defects will require new theory and modeling at longer length- and time-scales, and the development of new theoretical treatments to solve this problem largely depends on high-end computing.*

**Magnetic spin systems and single electron devices:** understanding the quantum level processes that govern spin and electron transport in new multilayer materials and ultra-small devices being investigated for the next paradigm in computer technology.

> *Active control and manipulation of the spin degrees of freedom in solid state devices requires detailed knowledge of electronic structure and quantum interactions, which in most cases will be determined only by extensive first principles calculations coupled with precise experimental investigations.*

Each of these opportunities presents a number of theoretical challenges, ranging from the formulation of new fundamental theory to the application of large-scale numerical calculations for predicting new phenomena and interpreting contemporary experiments. If you need the subcommittee to do so quickly, we can provide expanded descriptions of any of these opportunities, including more technical detail and a more complete picture of the scientific impact.

### *Connecting theory with experiment at the DOE facilities: Seizing new opportunities*

A key part of the scientific mission of the Department of Energy is the construction and operation of large experimental facilities for the scientific community. We have identified a need for stronger coupling of theory and computation to the experiments underway at these BES facilities as a major theme, expressed by both theorists as well as experimentalists in the community. Theory and computation could enhance scientific productivity in a strengthened partnership with experiment at these facilities in several ways. In areas where the experimental techniques and theoretical understanding are mature, theory and computation play the critical function of interpreting experiment, making it worthwhile to perform ever more detailed and complex investigations. In areas where the experimental methods are both novel and still rapidly evolving, theory can lead experiment, suggesting new areas of inquiry and proposing new kinds of experiments.

We find that at the existing light and neutron sources, for example, there appears to be little conscious or systematic effort by BES to stimulate and support, with targeted resources, theoretical partnerships with experimental efforts. We believe that this situation puts at risk DOE's ability to extract the maximum scientific benefit from those facilities. You may expect the subcommittee report to expand on this point.

At each of the nanoscience facilities presently under construction, on the other hand, we find an effort to build an accompanying theoretical and computational effort as part of the development of the facility. We applaud the fact that the leaders of these facilities have recognized that theoretical research is critical to the success of the nanoscience facilities. In public testimony we heard that in many cases, structures cannot be deduced and interpretation of nanoscience experiments is impossible without theoretical input and analysis. Without associated theory programs, the nanoscience facilities would be at risk of not realizing their full potential. The subcommittee report is likely to opine further on possible models for these theory programs and on the appropriate scale of both human and computational resources.

The subcommittee has not yet received enough testimony regarding the theory programs at the Spallation Neutron Source to begin to form recommendations. This facility, the most expensive of all the experimental facilities of the Office of Science, is focused on an array of scientific problems across chemistry and materials science, and for many of these problems mature theoretical and computational approaches already exist. The Office of Science thus has much to gain by partnering with new programs in theory and computation, both to interpret the results of planned experiments and to spark the design of new experiments.

Finally, we have found compelling evidence of the need for theory programs associated with the “fourth generation” light sources under construction and development at the DOE Laboratories. For these facilities theory and computation must at this stage play the role of leading and driving experiment. In addition, there are important opportunities for theoretical studies to inform and perhaps dictate the designs and specifications of the experimental facilities themselves. New facilities open up new domains of scientific inquiry, and we expect our report to discuss the need for theoretical efforts that can identify and propose new physical processes that will become accessible using ultrafast and ultra-intense light sources.

## *Supporting new styles of theory and computation in the BES portfolio – scientific codes as shared instruments*

The subcommittee report will discuss some of the changes that have occurred in the theory enterprise as a result of improvements in algorithms and the increased power of computers, and the opportunities that have been created by advances in computational capability at all levels, from small clusters, to supercomputers, to the recently announced leadership-scale computing facility.

One clear theme has emerged already from our deliberations. The success of the entire BES theory community has been and is being retarded by a lack of support for the development and maintenance of shared scientific software. Many of the computer codes that make it possible to express and exploit theory in all of the scientific areas in the BES portfolio are large-scale instruments in their own right. In particular, to exploit high-end parallel computing, like the leadership-class facilities being contemplated by the Office of Science, individual research groups or consortia must develop complex computer programs that frequently consist of tens or hundreds of thousands of lines of code. But with almost no exceptions, BES does not fund the maintenance, documentation, and evolution of those codes for the use of groups other than the one in which they were developed. These are demanding tasks that scientists cannot afford to do for one another as a form of philanthropy. Our discipline-specific scientific software in BES has been largely unshared, resulting in frequent and unnecessary duplication of effort. These potentially important tools are also rarely exploited by experimentalists. This situation is a tremendous loss for the scientific community.

In order to enhance the ability of BES theorists to take advantage of high-end computer architectures, there should also be opportunities for partnerships with applied mathematicians and computer scientists in the development of algorithms and software. This would be along the philosophical lines of the Scientific Discovery through Advanced Computing (SciDAC) program – a concept that appears to have considerable support in the community.

The subcommittee will recommend that BES consider supporting the development and maintenance of scientific codes in the disciplines in its portfolio, just as it now funds the development of shared beam lines at its experimental facilities. This type of project would complement the projects now supported in theory and computation by BES, and create community scientific software that would be shared and further developed by theoreticians and frequently used directly by the associated experimental communities. This style of support would distinguish BES support of the physical sciences from that of the other federal agencies, and it would be consistent with the mission of the Office of Science to create new scientific capabilities for the nation. Investments such as these will also be critical in allowing BES researchers to take full advantage of the capabilities of DOE's leadership-class computing facilities.

## *Conclusion*

The deliberations of the subcommittee on Theory and Computation have already identified exciting new scientific opportunities for BES and highlighted specific issues regarding the DOE experimental facilities and the support of theoretical research over the entire portfolio. However, the subcommittee, as it identifies new opportunities for investment, is keenly aware of the hazards of the potential competition between new investments in these areas and the current investments in the existing base research programs. It is our intention to provide the Office of Basic Energy Sciences with a compelling case for increased investment in theory and computation that can successfully support the argument for greater resources for BES.

Sincerely,

*The BESAC Subcommittee on Theory and Computation*


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