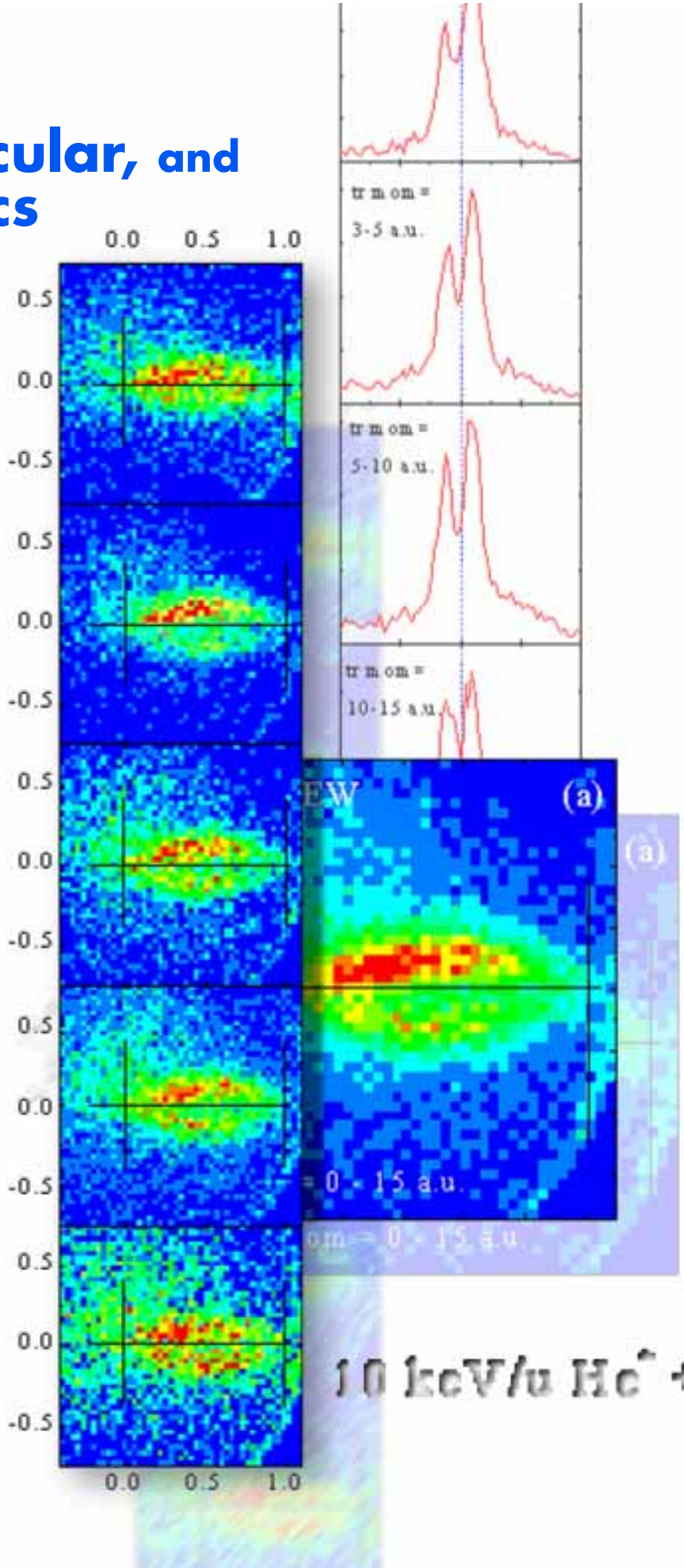
Atomic, Molecular, and Optical Physics Workshop

Final Report

of the workshop held
September 21-24, 1997
in
Chantilly, Virginia



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PREFACE

This document contains the final reports from the five panels that comprised a Workshop held to explore future directions, scientific impacts and technological connections of research in Atomic, Molecular and Optical Physics. This workshop was sponsored by the Department of Energy, Office of Basic Energy Sciences, Chemical Sciences Division and was held at the Westfields International Conference Center in Chantilly, Virginia on September 21-24, 1997. The workshop was chaired by Lloyd Armstrong, Jr., University of Southern California and the five panels focused on the following topics:

Panel A: Interactions of Atoms and Molecules with Photons - Low Field
Daniel Kleppner (Massachusetts Institute of Technology), chair

Panel B: Interactions of Atoms and Molecules with Photons - High Field
Phil Bucksbaum (University of Michigan), chair

Panel C: Surface Interactions with Photons, Electrons, Ions, Atoms and Molecules
J. Wayne Rabalais (University of Houston), chair

Panel D: Theory of Structure and Dynamics
Chris Greene (University of Colorado), chair

Panel E: Nano- and Mesocopic Structures
Paul Alivisatos (Lawrence Berkeley National Laboratory), chair

The choice of focus areas reflects areas of significant interest to DOE/BES but is clearly not intended to span all fields encompassed by the designation of atomic, molecular and optical physics, nor even all areas that would be considered for review and funding under DOE's AMOP program. In a similar vein, not all research that might be suggested under these topics in this report would be appropriate for consideration by DOE's AMOP program. The workshop format included overview presentations from each of the panel chairs, followed by an intensive series of panel discussion sessions held over a two-day period. The panels were comprised of scientists from the U. S. and abroad, many of whom are not supported by DOE's AMOP Program. This workshop was held in lieu of the customary "Contractors Meeting" held annually for those scientists currently receiving support from the DOE AMOP Program. The current DOE principal investigators attended the workshop, presented posters on their current research, and engaged in discussions with the panel members. Thus, the reports contained in this document reflect their active participation.

The workshop and this report are intended to stimulate and provoke the thinking of those interested in research in AMOP. Anyone who would like to submit a proposal (Grant Application) for consideration and review can do so at any time. This document as well as the annual Summaries of Research in the Chemical Sciences should be useful reference materials to consider. Both are available either as hard copy or on our website at:
http://www.er.doe.gov/production/bes/chm/chmhome.html

The web version of the FY 1997 summaries of research can be easily searched by several criteria including both scientific discipline and relevant technologies. Those considering submitting a proposal are encouraged to prepare a brief preproposal prior to preparing and submitting a full proposal. Preproposals can be submitted and discussed with staff in the Chemical Sciences Division. All relevant information, including phone numbers and e-mail addresses, for all Division staff is available on our website and in the hardcopy version of the research summaries. Preproposals should be addressed to the Division Director with copies to the appropriate Team Leader and Program Manager. This will ensure full and timely consideration.

Patricia Dehmer
Associate Director of Energy Research for the
Office of Basic Energy Sciences
U. S. Department of Energy

FORWARD

Recent years have seen a virtual explosion of activity and advances in the fields of atomic, molecular and optical physics. Development of new instrumentation and new experimental techniques has proceeded at an unusually rapid rate, leading to discovery of many new phenomena and much improved characterization of previously known phenomena. New theoretical approaches have enabled higher levels of understanding of old problems, helped to provide explanations for newly discovered phenomena, and pointed out directions for new exploration.

The Department of Energy Office of Basic Energy Sciences held a workshop on Atomic, Molecular, and Optical (AMO) Physics on September 21-24, 1997, to review advances in selected areas of AMO physics and to consider the impact of these changes on the closely allied fields of surface interactions and nanostructures. Emphasis was placed on identifying future directions in all of these areas that are ripe for exploitation. The papers that follow describe the conclusions of the AMO panels that reviewed high-field and low-field regimes and the theory of structure and dynamics, as well as of the panels that looked at surface interactions and nanostructures and their relations with AMO physics.

One of the central themes running through all of these reports is that many of these new developments enable or should in the future enable a great expansion in our ability to control broad classes of physical phenomena. Control of novel photon and atom states, of quantum dynamics of collisions and chemical bonds, of atoms on surfaces, of chaotic behavior in nonseparable dynamical systems, of size, shape and properties of nanostructures is increasingly possible. Control of these and other similar phenomena promises to open up new and exciting fields of research, and to lead to the creation of powerful new experimental and theoretical techniques for further exploration. Achieving this increased level of control and of the understanding of these complex systems that this control implies could enable very significant advances in many important areas of research. For example, understanding and control of the quantum dynamics of collisions and chemical bonds is fundamental to the control and manipulation of chemical reactions.

Another important interconnecting theme is that of the continuing rapid evolution of probes, in particular those that utilize light. Developments in lasers are leading rapidly to ever higher intensity, ultrafast pulses of defined shape, shorter wavelength, greater stability and narrower lines. Third generation synchrotron light sources are providing unprecedented resolution and sensitivity in the high-energy photon range. New approaches to the production of photon beams having nonclassical properties have the potential to enable spectroscopic sensitivity beyond the standard quantum limit. These and other new probes enable important new experiments not only in AMO research, but in many other fields supported by the Office of Basic Energy Sciences as well.

Several of the reports discuss recently created novel states of matter or matter and field. The ultimate utility and application of these states is unknown, but many seem to have great potential for significant impact. Bose Einstein condensates (BEC) are one of these. Already, researchers have been able to use BEC to create “atom lasers” that display phase coherence between beams of atoms, raising the possibility of, for example,

greatly increased capabilities in lithography. In other work, new states are being created in which matter and field form systems with unusual properties. Among these at one end of the energy spectrum are entangled states, created in micromaser cavities with a small number of photons and atoms; at the other end are states in which external field strengths are comparable to the strength of the binding fields of the matter.

The papers on nanostructures and surface physics clearly show the very significant impact of existing AMO technologies and theories on these fields. In addition, as workers in these fields look to the future, they find numerous examples where developing AMO technologies should be of considerable importance in enabling the fields to move ahead. This theme of AMO as an enabling science is found throughout all of the reports, with each paper discussing areas outside of AMO where new technologies, instrumentation and theories being developed to advance the physics of AMO might find applications. Numerous potential important enabling applications of AMO technologies to other areas of interest to DOE and in particular BES are indicated throughout the reports.

Since the emphasis of this meeting was on future directions, it is not surprising that there are many obstacles to full realization of the potential of most of the areas of research discussed in these reports. In many, such as the matter-field states in which external field and binding field strengths are comparable, there are serious theoretical and computational difficulties that must be overcome in order to achieve understanding and predictability. In others, the experimental techniques and instrumentation are just now reaching a level of sophistication that enables “proof of principle” experiments, and considerable development will be required before real investigations and exploitation of the field can occur.

Overall, the picture that emerged from this workshop was of fields that are extremely dynamic and exciting, and critical to the development of many other fields of science. Important new phenomena, new theories, new techniques, new understanding were to be found in all of the areas that were discussed. Development is moving at an unprecedented rate in many of these areas. The challenges, the opportunities, and the excitement were clearly described by the participants, and well captured by the contributions contained in this volume.

Lloyd Armstrong, Jr., Chair

Panel A: Interaction of Atoms and Molecules with Photons - Low Field

Chair:
Daniel Kleppner (Massachusetts Institute of Technology)

Panelists:
Uwe Becker (Fritz Haber Institut der MPG)
Nora Berrah (Western Michigan University)
William Cooke (College of William and Mary)
Robert Field (Massachusetts Institute of Technology)
Wallace Glab (Texas Tech University)
Wendell Hill (University of Maryland)
Eric Horsdal-Pedersen (Aarhus University)
Stephen Leone (University of Colorado, JILA)
Marsha Lester (University of Pennsylvania)
Stephen Lundeen (Colorado State University)
Cheuk-Yiu Ng (Iowa State University)
Erwin Poliakoff (Louisiana State University)
Stephen Pratt (Argonne National Laboratory)
William Stwalley (University of Connecticut)
Michael White (Brookhaven National Laboratory)

Outline Summary

1. Atomic and Molecular Manipulation and Control

2. Advanced Spectroscopies with New Light Sources

3. Quantum Dynamics of Interacting Systems

Summary

Laser cooling and trapping techniques, synchrotron light sources, advanced imaging methods and other innovations have opened frontiers for research in atomic and molecular physics. Opportunities encompass fundamental studies of structure and dynamics, the development of knowledge and data for other disciplines and for industry, and the development of new technologies. We describe opportunities in three categories.

1. Atomic and Molecular Manipulation and Control

Light force techniques make it possible to cool atoms to sub-microkelvin temperatures and to study ultra cold collisions and new types of molecular association. They have led to the creation of matter-wave optics and to Bose-Einstein condensation in an atomic gas. These developments have created opportunities for the study of Bose-Einstein condensates, the creation of new condensate systems, the development of atom lasers, and new studies of fundamental symmetries. The cooling methods may be extended to molecular systems to create new types of spectroscopy and studies of dynamical processes. Applications for the research exist in areas ranging from biology to nanotechnology. In addition, new techniques to control and manipulate ions, including ion storage rings, can be employed to study molecular ionic systems that are important in the atmosphere and in low energy plasmas.

2. Advanced Spectroscopies with New Light Sources

Two developments have stimulated the creation of new spectroscopies and new methods for the quantum control of atomic and molecular processes. These are the creation of third generation light sources and practical methods for generating ultrashort optical pulses. Research opportunities with third generation light sources include studies of the basic physics of photon-matter interactions, new investigations of the many-body problem, studies of the product distributions from photo-initiated dissociation, and studies of the structure and dynamics of highly charged ions. The studies are germane to problems ranging from the origin of radiation damage in cells to hot plasmas in stars. Ultrashort optical pulses can be tailored to study atomic and molecular wavepacket dynamics. Tailored pulses can be used to control molecular dynamics and the products of chemical reactions, and can provide new diagnostics for identifying atoms and molecules in hostile environments.

3. Quantum Dynamics of Interacting Systems

Understanding chemical reactivity requires understanding the quantum dynamics of interacting systems. Scattering, including photo scattering and photo ejection, is the chief experimental tool for studying quantum dynamics. The development of novel photon sources and time- and position-sensitive imaging and co-incidence methods opens the way to study the mechanisms of energy exchange within a molecule. Reactions between simple systems can be observed with a detail approaching complete quantum specification. There are opportunities for extending these experiments to more complex systems and for developing visualization techniques for interpreting the voluminous data. In addition to elucidating the basic processes of chemical reactivity, such studies are important to astrophysics, plasma physics and materials science.

1. ATOMIC AND MOLECULAR MANIPULATION AND CONTROL

1A. Introduction

The underlying goal for much of the research in atomic, molecular and optical physics (AMOP) is to understand atomic, molecular and optical processes with the ultimate clarity that quantum mechanics allows. Achieving this goal experimentally requires tools for preparing atomic and molecular samples in specific states, and for controlling their motion. Recently there has been a revolution in techniques for controlling not only the internal states of atoms but also their translational states, providing qualitatively new levels of control. These methods have opened frontiers of research, generated strong links to other areas of science, provided new opportunities for metrology, and are starting to have an impact on technology.

1B. Light Forces—Manipulating Matter by Light

Motion is an impediment to the study of atomic, molecular and ionic structure and dynamics. Motion blurs spectral lines and obscures structure. It limits observation time, degrades the control of collision kinetics and introduces undesirable complexities into observations. For this reason, experimental progress in AMOP has often been propelled by progress in controlling atomic and molecular motion. Early efforts to control motion employed effusive atomic beams and mechanical velocity selectors. Supersonic beams were developed that further reduced thermal effects in atoms and molecules, creating a new arena for studying atomic and molecular dynamics and also the science of clusters, including the discovery of the fullerenes—honored by the 1996 Nobel Prize in chemistry. In a parallel development, the creation of ion traps led to essentially total control of ionic motion and opened the way to advances in spectroscopy, collision dynamics, plasma processes, and metrology.

Within the past decade a revolution in atomic control has been precipitated by the development of light force techniques. The significance of this revolution was dramatized by the award of the 1997 Nobel Prize in physics for the development of atomic light force methods. The essence of the advance is this: In the past, translational motion had to be treated classically, and kinetic energy was unquantized. Today, the ultimate limit in translational control has been achieved; it is now possible to put atoms into a single translational quantum state and to observe their behavior in complete detail.

The new methods employ laser techniques for slowing, cooling, and manipulating atoms, and optical and magnetic traps for confining them. The traps can hold large numbers of atoms at sub-microkelvin temperatures. These methods make it possible to witness the photo-association of free atoms into unusual molecular states; to study ultracold collisions in which the dynamics can be controlled by electric, magnetic, and laser fields; to observe phenomena such as Bloch oscillations—collective oscillations in a periodic potential—and to realize the quantum kicked rotor—a paradigm in the study of nonlinear dynamics.

The observation of Bose-Einstein condensation (BEC) in an atomic gas is the most dramatic achievement of the light-force techniques. The clarity with which the Bose condensates can be observed and the exquisite control by which they can be manipulated, as well as the novelty of the phenomena, have generated a new field of study that has attracted enormous interest in AMOP by condensed matter scientists, chemists, and others. The phase transition, the collective motions and transport and relaxation processes of the condensate, have all been studied. A study of the phase coherence of the condensate resulted in the creation of an atom laser.

Closely related to these developments is the creation of the field of matter wave optics. Several types of atom-wave interferometers have been demonstrated and optical elements such as mirrors, lenses, gratings, beam splitters, and hollow guiding fibers, have been developed. The interferometers have been employed to measure atomic and molecular properties, to investigate the decoherence in quantum systems, to measure fundamental constants, and to create ultrasensitive accelerometers. Atom-optics methods are being applied to new types of lithography, and have already been used to create length standards for scanning microscopies.

Scientific opportunities and applications include:

- Addressing fundamental questions on BEC including transport, condensate dynamics, coherent collisions, compound condensates, and interactions with light. The condensates may make possible new studies in quantum entanglement.
- Exploring new condensates and developing methods for increasing the size of BEC condensates. Such methods could be applied to much more intense atom lasers.
- Developing the atom laser as a scientific tool, and exploring its applications to nano-technology.
- Extending photoassociative spectroscopy of submillikelvin atoms to study long range molecular states of other atoms (for instance the hyperfine-free alkaline earths, and metastable rare gases) and also to study atom-molecule and molecule-molecule photoassociation. Such studies provide parameters such as scattering lengths, dissociation energies, and spin-flip cross sections. These are critical for understanding very low temperature dynamics and BEC.
- Developing light force and trapping techniques for molecules. Trapped molecule methods would provide new ways to study molecular structure, chemical reactions, and interatomic and inter- molecular interactions that may ultimately yield BEC of state-selected molecules, and molecule lasers.
- Applying low temperature molecular spectroscopy to new studies in molecular and biological physics. For example, low frequency torsional modes in biopolymers can be cooled to ultralow temperatures, perhaps yielding new insights into protein folding.
- Developing light force techniques for other applications. A notable example is the early development of optical tweezers, which are used in polymer studies, biology, and other applications. These tweezers are able for the first time to directly manipulate matter within a living cell and to stretch and even break an individual polymer or DNA molecule.

1C. Fundamental Studies

Closely related in spirit to the study of light forces is a body of research in AMOP centered on the study of fundamental symmetries and other fundamental phenomena. The goal is to deepen our basic understanding of physics, but in accomplishing this the research has often pushed forward the frontiers of experimental control, and of metrology.

Among the most precise studies in AMOP are a series of experiments on parity nonconservation (PNC) effects in atoms. These have contributed new information to the Standard Model and they have led to the discovery of a new type of nuclear current—the anapole moment. Laser power buildup cavities for these experiments have advanced laser mirror technology in developments that led to cavity ring-down

spectroscopy and that have been taken up by industry. New opportunities for this line of research have been opened by advances in light force techniques and in the trapping of radioactive atoms.

Atomic studies of the fundamental symmetries probe the basic forces that govern the interactions in nature and search for new types of fundamental particles. Such investigations range from the extremely high precision spectroscopy of neutral and ionic atoms and molecules to the creation of exotic atoms and antimatter. These experiments generally push the frontiers of experimental techniques and are a source of new technology for other sciences and for applications. Studies of QED in atoms are spurring methods to measure optical frequencies that have potential applications in communication and control. Searches for electric dipole moments of fundamental particles have led to the development of magnetometers and improved methods for measuring and controlling electric and magnetic fields. Investigations of antimatter atoms have been instrumental in the development of ion trapping.

In a different domain, new types of light have been created with unique quantum properties. These include squeezed light, correlated photon beams, and quantum solitons. A common feature is that the fluctuations lie below the so-called standard quantum limit associated with the vacuum state of the electromagnetic field. In addition to their fundamental interest, such nonclassical light sources may have spectroscopic applications. Nonclassical light has enhanced spectroscopic sensitivity beyond the standard quantum limit. If this technique can be made practical, it offers opportunities for advances in ultrasensitive detection and it may increase the sensitivity of gravity wave detectors such as LIGO. Closely related to these developments are the creation of new types of photon and atomic states using the methods of cavity quantum electrodynamics.

Opportunities include:

- Exploiting atom trapping and cooling techniques for tests of PNC interactions and other fundamental tests.
- Developing new methods for the spectroscopy of short lived atomic species.
- Applying optical methods to polarize atomic nuclei to create polarized beams and targets for nuclear and particle physics. Recent studies on the spin structure of the neutron through electron scattering from polarized helium-3 nuclei provide a notable example.
- Finding more efficient methods for polarizing rare gas nuclei. The production of these nuclei have led to the development of an important new medical diagnostic technology—magnetic resonance imaging with polarized rare gases—with greatly enhanced sensitivity and resolution.
- Investigating the behavior of atomic systems in nonclassical light fields to understand altered lineshapes and decay rates, nonlinear mixing, and new types of lasing behavior.
- Studying entangled atomic states using the methods of cavity quantum electrodynamics to obtain new techniques with potential applications in the general area of quantum computing and quantum cryptography.

1D. Molecular Control

Molecular spectra can be vastly simplified, and new types of molecular states accessed by methods such as multiple optical-optical and optical-RF resonance techniques, polarization labeling spectroscopy, multiphoton ionization, laser-induced fluorescence, and stimulated Raman adiabatic rapid passage. Beyond these, molecular spectra can be greatly simplified by cooling the molecules. Spectroscopy has been carried out to temperatures as low as 1K using seeded

supersonic molecular beams or solid rare gas matrices. More recently, temperatures as low as 0.4K have been achieved, for example by studying species in helium-4 clusters.

Opportunities include:

- Extending methods for manipulating and controlling the vibrational populations of molecules to their rotational and electronic states. Such control would provide new pathways to the studies of collision dynamics and chemical reactions.
- Developing methods for cooling and trapping molecules. These methods could provide new levels of resolution and vastly simplify molecular spectroscopy and provide a new arena for the study of collisions and reactions.
- Controlling detrimental loss processes in traps by controlling collision dynamics.

1E. The Control of Ions

Ions can be manipulated and controlled with traps for collision studies, ultrahigh precision spectroscopy, studies of ion crystals and strongly coupled plasmas, and for applications to quantum logic gates and quantum computing. Ion trap methods have also been used to observe vibrational relaxation of molecular ions, and dissociative and dielectronic recombination at very low energies. Because dissociative recombination can play an essential role in determining the distributions of free radicals in low energy plasmas, this research can have important applications in materials processing and etching.

In heavy ion storage rings molecular ions can be stored at MeV energies for several seconds, allowing them to cool rotationally and vibrationally. Reaction products from dissociative recombination and branching ratios for electronic states can be identified using imaging techniques. Ions that can be conveniently studied include O_2^+ and N_2^+, which are relevant to the physics and chemistry of the Earth's atmospheres (N_2^+ is also important on Mars), and H_2^+, which is important to the chemistry of the interstellar space.

Opportunities include:

- Producing state-selected molecular ions directly from ultracold atoms by photoassociative ionization.
- Manipulating ions in storage rings by laser light, providing a new control over reaction dynamics.

2. ADVANCED SPECTROSCOPIES WITH NEW LIGHT SOURCES

Two developments are stimulating the creation of new forms of spectroscopy and new methods for the quantum control of atomic and molecular processes. These are the creation of third generation light sources to extend the power and resolution of spectroscopy in the UV and short x-ray regimes, and the development of practical methods for generating tailored ultrashort optical pulses. Additional opportunities due to free electron laser light sources are discussed by the Panel on Interactions of Atoms and Molecules with Photons: High Energy.

2A. Third Generation Synchrotron Sources

The photon beams now available from third generation synchrotron light sources developed at Berkeley (ALS) and Argonne (APS) offer new opportunities to study radiation-matter interactions that require very high energies, spatial resolution, spectral resolution, or polarization. The low emittance of third generation storage rings makes it possible to employ undulator or wiggler devices that can provide radiation into the hard x-ray region with unprecedented intensities and spatial collimation. In atomic and molecular physics,

these developments open new avenues for studying multi-electron excitation and inner-shell photoionization. For example, high brightness undulators in the UV can be used for "universal" detection of neutral product species via one-photon ionization with sensitivities rivaling conventional electron-impact sources, but without the associated difficulties of unwanted dissociative ionization and a hot source region.

Complex atomic, molecular and ionic processes can be studied by combining third generation synchrotron radiation with high resolution detection techniques that simultaneously detect and analyze every possible outcome channel. These methods also make it possible to image the fragments emitted in photoprocesses-ions, electrons, neutral, and photons—and to detect their coincidences. Because polyatomic molecular species can exit in different chemical isomers with distinctly different chemical properties and energetics, the detailed understanding of photodissociation, electron dissociation, and reaction processes is possible only with such an identification of nascent product chemical structures.

Opportunities include:

- Testing the basic assumptions that have been used traditionally to interpret photon-matter interactions; for instance, the independent particle, impulse, and dipole approximations.
- Conducting quantum mechanically complete experiments that determine the alignment and orientation parameters for atomic and molecular targets, using circularly or elliptically polarized light from helical undulators.
- Advancing our understanding of the many-body problem, one of the central problems in atomic, nuclear and condensed matter physics, through negative ion spectroscopy, as suggested by theoretical studies.
- Identifying nascent product distributions of atomic and molecular fragments, resulting from reactive scattering, electron attachment, and photo-initiated dissociation, ionization, detachment and desorption.
- Studying endothermic reactions with low cross sections and other processes that result in difficult to detect, low-yield neutral products.
- Combining ultra-high resolution capabilities of small aperture monochromators with the beams from undulators to produce narrow-band UV and soft x-ray radiation to study photoionization of atoms, molecules, clusters and ions, possibly with complete quantum state specification and kinematic imaging.
- Employing high resolution photoelectron-photoion coincidence techniques, to prepare atomic and molecular ions with specified quantum states for scattering and spectroscopic experiments. Neutral atoms (O, N, and C, etc.) and radicals (OH, CN, CH_3, and C_2H, etc.) can be prepared by photodissociation of appropriate precursor molecules using the high flux VUV synchrotron radiation.
- Employing light from third generation synchrotron sources to create new species; for example, "hollow" atoms in which the core electrons are simultaneously excited to high-lying states. Such atoms provide a new arena for observing many-electron systems and testing theories of highly correlated electron motion.
- Studying the structure and dynamics of highly charged ions by combining radiation from third generation synchrotron sources with targets of highly charged ions. These studies will provide precision data on ionic structure and electron-correlation effects, benchmarks for theoretical studies of multielectron interactions and data for plasma modeling and ion-beam lithography.

2A-2. Related Science and Applications

X-ray absorption spectroscopy with third-generation synchrotrons can provide element-specific and chemical state-specific detection of trace contaminants in biological materials, soils and groundwater. The spectroscopy can be applied to understand radiation damage in complex molecules such as DNA, for the damage originates in core excitations that are fundamentally atomic processes. A real understanding of radiation damage is possible only through studies of the complex decay processes in isolated atoms and simple molecules.

Studies in the structure and dynamics of highly charged ions is vital for understanding hot plasmas in stars, in fusion reactors, and also in x-ray lasers.

The energetic, spectroscopic and dynamical information from molecular studies with third generation synchrotron sources can be important to combustion chemistry, plasma science, and environmental and atmospheric chemistry. For instance, cation spectroscopic studies can yield invaluable spectroscopic and geometric information on the neutral ground state. Rotationally resolved photoionization and photoelectron spectroscopic studies of reactive intermediates, such as CH, CH_2, NH, NH_2, HCO, CH_3O, and C_2H, are important in combustion, plasmas, and atmospheric chemistry.

2B. Ultrafast Lasers and Wave Packet Dynamics

Sub-picosecond laser systems have become practical laboratory instruments. Their development has generated an explosion of new research on the control of elementary photochemical and photophysical processes by using sequences of ultrafast pulses with precisely shaped spectral and temporal profiles. The pulses, which are shorter than the period of typical molecular vibrations and rotations, or the time between collisions in a dense gas, make possible new ways for manipulating and interrogating the internal dynamics of molecules. Several groups have employed shaped optical pulses to generate atomic and molecular wave-packets. These techniques may be able to provide such precise control over internal interactions that the final products of chemical reactions can be selected by suitably arranging a sequence of shaped pulses. In related control experiments, interference between two optical excitation pathways has been used to control the branching ratios between photoionization and photodissociation in hydrogen iodide, and interference between one- and two-photon excitations has been used to control the photoionization angular distribution and the direction of the flow of current in solid state devices.

In a somewhat different context, new types of wave-packets have been created in Rydberg atoms by employing combinations of laser light, microwave fields and time-varying electric and magnetic fields. The electronic current can be stationary and highly localized, as in a “circular” Rydberg state, and the electron can even be localized in a stationary wave packet, as in a coherent elliptic state. Such atomic states have already been employed in studies of quantum optics, high resolution spectroscopy, and scattering. They have also been employed to study the physics of the semiclassical regime, a problem which is of deep interest in molecular physics where semiclassical approximations are often appropriate.

Opportunities include:

- Extending these tailored pulse techniques by combining several shaped, multicolor pulses.
- Investigating wave packet dynamics by experimentally executing algorithms created with optimal control theory.

- Employing short-pulsed, multi-step excitation to create a new collision-free tool for identifying atoms or molecules, even in hostile environments such as plasmas or flames.
- Investigating the possibility of preparing species in wave packets to control the dynamics of interactions, i.e., of controlling molecular reactions.
- Developing new methods for creating and studying the physics of localized atomic states, and for applying these states to scattering problems and studies of semiclassical physics.

3. QUANTUM DYNAMICS OF INTERACTING SYSTEMS

Understanding the fundamentals of chemical reactivity demands understanding the quantum dynamics of interacting systems. Such an understanding would contribute not only to fundamental chemical dynamics but to essentially every area of chemistry and related disciplines such as atmospheric science, combustion, astrophysics, plasmas, and materials science. The quantum dynamics of interacting systems holds the key to controlling and manipulating reactions. It is difficult to overestimate the power that such control would provide.

Just as spectroscopy is the chief experimental tool for studying the structure of atomic and molecular systems, scattering—including photon scattering and half-scattering (photo ejection processes)—is the primary experimental tool for studying dynamics at the quantum level. Spectroscopy is also an important tool for studying dynamical processes such as photoassociation and photoionization, and it can provide essential diagnostics for scattering experiments. And just as spectroscopy has undergone a revolution, so too has scattering, for the clarity with which scattering events can be observed, the variety of systems that have become accessible, and the range in dynamical variables with which they can be studied, have expanded enormously.

3A. Complete Scattering Experiments

As its name suggests, a complete scattering experiment provides all the information about a scattering event that quantum dynamics allows us to know. Instead of an average transition rate, one talks of the quantum mechanical amplitude for passing from one quantum state of the collision partners to another. Such an experiment requires precise measurement and control of the incoming and outgoing momenta of the scattering partners, and also their polarization, alignment, and orientation. With such measurements one can obtain both the magnitudes and phases of the scattering amplitudes for targets and projectiles such as atoms, ions, photons, electrons, and simple molecules.

Although a complete scattering experiment remains an ideal, even in cases where it cannot be fully achieved the concept can nevertheless be employed to distinguish the important degrees of freedom in complex systems. Techniques employed in complete scattering experiments include polarized beams and targets, spin resolved electron detection, and laser-selected molecular state preparation and detection. The research has been propelled by the recent development of time- and position-sensitive imaging methods that provide what are known as 4π detection, detectors in which every scattering partner in a collision is observed and fully measured. Their high collection efficiency can make coincidence experiments feasible. These detectors have been applied to photo fragmentation (“half collisions”) and particle-induced reactions involving atoms and simple molecules, for instance, to the photodissociation of molecular hydrogen. Other advances include photofragment imaging and multiphoton ionization of neutral products.

Scientific opportunities and applications include:

- Extending complete scattering studies to more complex systems such as polyatomic molecules, radicals and ions. These experiments, with their full specification of initial and final quantum states, can reveal the mechanisms for energy exchange between electronic, vibrational, rotational and translational degrees of freedom. Such experiments have been carried out on reactions between atoms and simple diatomic molecules, for instance deuterium and molecular hydrogen, and on the competition between autoionization and predissociation in highly excited small molecules.
- Extending these techniques to clusters. Collision experiments with complete specificity are not yet possible due to the high density of final quantum states in a cluster, but much can be learned from collisions with electrons, atoms, and ions, in experiments employing mass spectrometry and time-of-flight energy analysis. Such experiments can reveal, for instance, the mass distribution of photofragments, information that is essential for understanding cluster-cluster and cluster-surface interactions.

3B. Visualization of Quantum States and Dynamics

A serious problem in interpreting data from complete scattering experiments and comparing the results with theory is that even relatively simple systems require complex multidimensional representations. It is essential to find methods to view this information in ways that yield insights into the essential degrees of freedom, time scales, and the various competing mechanisms. The particular way in which the results are presented is crucial in determining whether or not the essential processes can even be recognized.

In some cases, a subset of the coordinates can be employed in a reduced dimension visualization scheme. An example is the femto-chemists' picture of a wave packet moving on a potential surface. This sort of picture aids in the design of experiments and in the identification of physically plausible coupling mechanisms. Non-separable, nonperturbative systems are often separable for a short time after a short pulse preparation of a localized excitation. In some cases the first stages of an interaction can be separated into active and passive degrees of freedom. In such cases the reduced dimension representation of the evolving system can show the evolution within a system of a well understood subsystem that has been previously studied in isolation.

In high resolution frequency domain spectroscopy, a spectrum can consist of entangled patterns arising from different values of a conserved observable. By comparing different spectra in which the entangled patterns appear with different relative weights, the dynamically independent pieces of the spectrum may be separated. For example, data on threshold double ionization of helium presented in terms of the momentum vector of the relative motion of the two electrons revealed a simple dynamical pattern that was essentially invisible when presented in terms of the conventional laboratory frame momentum vectors.

Scientific opportunities include:

- Developing reduced-dimension visualization methods for optimally representing a complex system in terms of weakly interacting subsystems. It is essential to find when such representation are appropriate, for they can introduce major simplifications.
- Developing pattern recognition techniques that can recognize an unspecified and unknown patterns in a large, high quality data set.

- Developing methods that reveal correlations in the internal relative momenta in multi-dimensional velocity space. Such methods are needed for understanding experiments in which the momentum vectors of all final reaction products are determined individually event by event.
- Investigating the use of collective, Jacobi, or other appropriate internal momentum coordinates, to reveal patterns that reveal the roles of two-body or collective interactions.

3C. Cross-Disciplinary Connections

Advancing our understanding of the quantum mechanics of interacting systems has broad ramifications and the impact of the research is not easily summarized. Nevertheless, it is of value to note some of these.

a. Comprehensive sets of dynamical parameters of atomic collision and half-collision processes provide information needed to derive partial cross-sections, angular distributions, and spin parameters of a large variety of atoms and molecules that are important in astrophysics, plasma physics, and materials science.

b. The interaction of highly charged atomic ions with atomic, molecular, electronic and ionic targets occupies a special place in the study of quantum dynamics of interacting systems because of their importance in high temperature plasmas. Such plasmas occur in such fusion plasma devices and laser-produced plasmas in both ICF and x-ray laser work. Opacities, charge-state equilibria and equations of state of such plasmas, as well as energy flow rates through short lived plasmas, can be calculated if relevant recombination, electron transfer, and ionization rates are known. Interactions involving ions in states appropriate to plasmas with electronic temperatures of a few hundred eV to over 100 keV are particularly important. In addition, a quantitative understanding of binary ion-ion and photon-ion interactions is needed for numerous plasma diagnostic techniques.

c. Understanding theoretically and experimentally the transition of scattering characteristics as a function of collision energy where the collision evolves from molecular characteristics (e.g., molecular wave functions, potential curves, etc.) to that of proceeding via atomic characteristics (e.g., impulsive interactions, atomic states, etc.) is important to basic physics and for applications.

Panel B: Interactions of Atoms and Molecules with Photon - High Field

Chair:
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Panelists:
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I. Executive Summary

In "high-field" interactions applied field energies equal or exceed the energy density of matter, so that the field controls the shape and dynamics of atoms, molecules, or other material systems. Using modern tools of high-field and ultrafast physics, one no longer merely observes nature, but can reshape and redirect atoms, molecules, particles, or radiation. This new drive towards harnessing quantum dynamics is enormously important to future developments in fundamental physics and applied energy science.

This document explores some of the ramifications and future fertile directions in this field. We divide the paper into three "grand visions," which are broad interconnected areas of AMO science. We begin with an executive summary:

I.A. Frontiers of Time, Intensity, and Wavelength

The first vision is the technical frontier of high fields and ultrafast optical pulses. The lasers that produce the intense fields are themselves designed and built using the principles of this new science of manipulating matter.

The past decade has seen a thousand-fold improvement in the peak power of solid-state lasers, and the future holds challenges for scaling to higher power with both solid-state and proposed free-electron sources. This progress will enable a wide range of science: new methods for more efficient inertial confinement fusion; compact laser-driven electron accelerators; sources of femtosecond (10^{-15} seconds) gamma radiation are some of the prospects.

Pulses will also become shorter, opening new areas of scientific inquiry previously undreamed. Femtosecond pulses, which were frontier research projects ten years ago, have been commercialized. Attosecond (10^{-18} seconds) pulses are actively investigated, and their control and measurement is a major challenge. Attosecond science will certainly center on the dynamics of atoms and molecules, since the time for scattering between a relativistic particle and an atom is on this timescale.

High-field laser-atom interactions have recently yielded intense coherent radiation at wavelengths as short as 27 Angstroms, which is short enough for biological imaging of femtosecond dynamics in the "water window." These light sources are major complements to synchrotron sources, delivering shorter pulses and greater control over coherence, on platforms that can be as small as a single table top. Still shorter wavelength sources have been demonstrated by scattering high intensity laser pulses from relativistic electon beams, or gating synchrotron radiation using laser-driven material switches. These advances bring kilovolt, megavolt, or even gigavolt photons into reach for laser-based time-resolved science of materials, chemistry, atomic physics, and possibly even particle physics.

We now control not only the wavelength, intensity, and duration of intense fields, but also their detailed shape, often down to the single-cycle limit. Pulse shaping is a major enabling element of the second "grand vision," which we call:

I.B. Harnessing Quantum Dynamics

Quantum dynamics in an atom or a molecular bond involves atomic-size fields (volts/angstrom), times (picoseconds and below), and frequencies (terahertz [10^{12} hertz] to exahertz [10^{18} hertz]). Our challenge is to achieve the technical ability to manipulate these states, the knowledge of how to design new "sculpted" states, and the wisdom of which new states will best accomplish our goals of new insight and useful applications.

Some of the frontier areas are fundamental, such as nonclassical properties of entangled states of the fields and matter, or the control of chaotic behavior in nonseparable dynamical systems. Others challenges lie at the interface between basic knowledge and technology. For example, adaptive feedback could

permit the newly formed quantum state to "teach" the external field how to alter its own shape. Sometimes this adaptive feedback is built into the quantum system, such as in relativistic plasma channeling or electromagnetically-induced transparency. An even more exciting vision for the future, however, is using fast computers and pulse shapers to alter intense fields according to instructions given by the quantum state, in order to optimize the state for a particular purpose. Some of the possible challenging applications of field-sculpted matter are:

- Bond-selective chemistry, in which a strong programmed optical field creates a rovibrational or electronic wavepacket in a molecule, which induces a specific change in unimolecular chemistry;
- Coherent collisions, where target or projectile are prepared in wavepacket states to control specific inelastic channels or select impact parameters;
- Quantum encoding, the encrypting of information into the amplitude and phase of nonstationary states. A strong programmed field pulse can key the return of the information as a quantum echo;
- Altering bulk properties, to control Bragg scattering, switch crystal domains, or move charge in solids and plasmas;
- Energy channeling, the controlled transport and deposition of laser energy into media which are generally opaque to weak or unshaped optical pulses. The mechanisms for energy channeling change dramatically when the intensity is very high, and this leads us to the third grand vision:

I.C. Matter in extreme conditions

At the extreme limit of high intensity in a laser focus, the electric fields can exceed trillions of volts per centimeter; magnetic fields can approach gigagauss; pressures aproach a gigabar. Physics in this environment is largely unexplored.

The high intensity can be converted to high pressure in solid-density targets, to produce conditions usually only observed in astrophysical objects or in thermonuclear reactions. Possible applications are advanced x-ray sources, or creating initial conditions for inertial confinement fusion.

New nonlinear effects occur when the work done on an electron during one optical cycle exceeds the electron rest mass (intensities exceeding one exawatt per square centimeter). Optical propagation in plasmas becomes dominated by the oscillating relativistic mass of the electrons. This coherent motion creates extreme charge-density gradients in plasmas, which can support fields of gigavolts per centimeter. Nonlinear Compton scattering in this regime creates harmonics from the relativistic motion of the electrons. Photoionization into such a relativistic continuum has not been studied.

At still higher fields, near the material limits for focused laser light in the laboratory but well beyond what has been demonstrated, electrons can be driven so violently that they emit electron-positron pairs through the process of coherent nonlinear Compton scattering.

Still higher intensities can be achieved in the center of momentum frame by back-scattering laser light from a relativistic electron beam, such as the SLAC linac. In the CM frame of the backscattered gamma ray and a counter-propagating laser beam, the field can rise to above ten million gigavolts per centimeter, at which field the vacuum itself becomes unstable, decaying into an electron-positron plasma. Positrons from this mechanism were recently observed at SLAC (D.L. Burke, et al., PRL 79, 1626 (1997)). This was the first example of coherent sparking of the vacuum.

I.D. Reading List and References.

This document was prepared at an intense two-day workshop in Chantilly, VA. As such, it can only provide qualitative and general information on the subject. There are, however, several important focused conferences in this field where current work

and future prospects are discussed in detail, and the reader may find additional details and documentation in their proceedings:

- Super-intense laser-atom physics (SILAP), most current meeting was in Russia in August, 1996. Proceedings edited by H.G. Muller and M. Federov.
- OSA Topical Meeting on Short Wavelengths and High Fields, most current meeting in Santa Fe in March, 1997, Proceedings edited by L. DiMauro and M. Murnane, not yet published. The previous meeting in this series is available: "High Field Interactions and Short Wavelength Generation, AUG 1994, St. Malo, France, Paperback, ISBN 1-55752-361-4, Available from the Optical Society of America.
- International Conference on Multiphoton Physics (ICOMP), most recent meeting in Munich in October, 1996, Proceedings edited by P. Lambropoulos.
- 10th ICFA Beam Dynamics Panel Workshop; 4th Generation Light Sources, Grenoble, France Jan 22-25, 1996, editor J.L. Laclare. The report is available on the web: http://www.esrf.fr/conferences/conclusions/webconc.htm
- Proceedings of the Seventh Advanced Accelerator Concepts Workshop, Lake Tahoe, CA, Oct., 1996, W. Chattopadhyay, J. McCullough and P. Dahl, Editors, AIP, conference Proceedings 398 (American Institute of Physics, New York, 1997).
- OSA Topical Meeting on Ultrafast Processes, ULTRAFAST PHENOMENA, MAY 1996, San Diego, CA, ISBN 1-55752-441-6, available from the Optical Society of America.
- University of California at Berkeley Fast Ignitor Physics Workshop, March 1997. Unpublished proceedings might be available from Dick Lee at Lawrence Livermore National Lab.

II. Opportunities in Intense-Field AMO Physics

Under high-field conditions, systems are driven by external fields to evolve more rapidly than their natural dynamical time scale. In quantum mechanical language appropriate to atomic and molecular physics, we state this as follows: A dynamical system u(t) evolves according to a Hamiltonian

$$(H + H'(t))u(t) = i\,\hbar\,[d(u(t))]/dt$$

Where H is the Hamiltonian of the system excluding the field interaction, and H' is the interaction hamiltonian. When H' evolves on a time scale faster than the natural dynamical timescale of u(t), and/or has a magnitude comparable to or greater than H, then we are in the high field regime. Typically, traditional approaches to dynamics in quantum systems such as perturbation theory do not reveal the physics of such systems.

Instead, the field and the matter form together a new system which generally has altogether new properties. Some of these properties will be critical for advances in energy science and technology, and these areas form the grand challenges of this field.

II. A. Frontiers of Time, Intensity, and Wavelength

In this section we will discuss the physical frontiers in materials, concepts, and optics at high fields, short wavelengths, and short pulsewidths. The generation of ultrahigh fields continues to go hand in hand with the generation of ultrashort pulses. The current state-of-the-art in solid state lasers operating at wavelengths near 1000-nm have yielded pulses as short as 5 fs and intensities 10^{22} W/cm^2. Currently, the intensity is limited primarily by the saturation intensity and geometry of the gain medium. The pulse duration is close to the limit of the uncertainty principle with a 5 fs pulse at 1000 nm consisting of approximately 3 optical cycles. If future technology could scale the current 1 cm diameter solid state lasers to 1 m, then intensities of 10^{26} W/cm^2 and concomitant field strengths of 10^{14} V/cm should be achievable.

If these technological extremes (e.g., scaling the size of gain medium) are achieved, where will the next advances come from? Laboratory intensities even above the solid state laser intensity limit of 10^{26} W/cm^2 might be reached by future Free-Electron Laser sources. Alternatively, the problem of scaling can be sidestepped altogether by the use of relativistic targets, which see the optical fields upshifted by a factor of gamma. Although many processes cannot be studied this way, relativistic electron and ion beams have already been used to study special regimes in high-intensity physics.

In the area of ultrashort pulse generation, if we push the limits of high harmonic generation for example to the 1000th harmonic of a solid state laser to 10-Angstroms (~ 1 keV) and assumed we could phase match this process then we would predict pulses of order 10 as (attoseconds). We have always inferred that the timescale for electron orbits was on this order, but we may soon have the capability to directly measure it and predict new phenomena!

High intensity ultrashort pulses are necessary to generate coherent sources of x-rays. Recent work by two groups have demonstrated coherent high harmonic generation within the water transparency window of critical importance for imaging biological processes. Among the many uses for this unique source of light would be the following: x-ray holography for medical imaging applications; table-top sources could complement synchrotrons as the sources of light necessary for x-ray lithography to push the limits below 0.1-micron line rules. Even shorter wavelength coherent radiation is proposed using high energy free-electron lasers. One such proposal would use the SLAC linac to generate 1-Angstrom level or ~ 10 keV photons. Beyond this, ultrashort pulses of gamma rays can be produced by backscattering intense lasers from relativistic electron beams. Photon energies up to 29 GeV have been demonstrated using this technique, opening the prospect of ultrafast nuclear or high energy science!

II.A.2. Frontiers and Physical Limits of Solid State Lasers

II.A.2.1 Laser-Based Mechanisms

Electromagnetic fields are proportional to the square root of the intensity in the focus of a coherent source. They are thus limited by the laser power (W) and the focal size, which, in turn, depends on the wavelength through diffraction limits.

The laser power is theoretically limited by the saturation fluence and the pulse length, both being ultimately determined by material constants (cross sections and bandwidth). In practice, additional properties like spectral absorption, thermal conductivity, temperature- and intensity-dependent index of refraction play a crucial role, limiting the number of suitable high-intensity laser materials to a handful of host materials, glasses and crystals. They typically contain a small concentration of ions with mostly vibrationally broadened transitions in the near-infrared as active components (solid state high power lasers).

Extensive and systematic screening of possible new materials over the last 5 to 10 years has yielded limited success in pushing the theoretical physical limits of intensity and field strength. We note that at least the bandwidth of the best known materials (like Ti:sapphire) is already within a substantial fraction of the photon energy, which is the ultimate physical limit, and can only be substantially improved by going to much shorter emission wavelengths.

The physical limits of laser field strength and intensity were increased by more than four orders of magnitude through introduction of chirped pulse amplification (CPA). We assume that this technique and the properties of presently known materials will essentially determine the maximum achievable intensities and field strengths for solid state high intensity lasers over the next 5 to 10 years.

Under this assumption, the maximum power out of 1 cm beam diameter of a solid state laser is between

100TW and 1 PW, the intensity (at wavelengths around 1um and nearly diffraction limited) is of the order of 10^{22} W/cm^2, the electric field strength is of the order of 10^{12} V/cm (1000 a.u.). A beam diameter (limited by materials processing) of the order of one meter would increase the focused intensity by another four orders of magnitude (10^{26} W/cm^2), and the field strength by two. However, we do not believe that this is a realistic assumption. Furthermore, even to reach the maximum values for 1 cm beam diameter requires enormous technological advances in laser etchnology (B- integral, stretching factors etc.). Hence, even for the visionary purposes of the present study we anticipate the following frontiers:

Laser power:	10 petawatts (10^{16} watts)
pulse duration:	5 fs
wavelength:	0.8 to 1 μm
Intensity:	10^{23} W/cm^2
Electric field strength:	10^{13} V/cm (2000 a.u.)
Magnetic field strength:	10^6 Tesla
ponderomotive energy:	~ 10 GeV (Relativistic scaling)

II.A.2.2 Other Mechanisms to Create Ultra-High Fields

The upper limits of the electromagnetic field strength as well as the lower limit for pulse durations will be encountered in relativistic heavy ion collisions. At the LHC at CERN, Pb^{82+} ions at energies of ~3000 GeV/nucleon will collide head-on and generate electric field strengths exceeding 10^{23} V/cm for pulse durations of only 10^{-28} s.

II.A.3. VUV- and XUV- Sources for High Intensities and High Fields

The physical limitations of present technology invokes the need for developments beyond our conventional approach for generating coherent high fields and ultra-short pulses at short wavelengths. The issue is an old problem associated with materials, e.g. absorption, as one pushes the wavelength beyond the VUV region. The options are either the use of nonlinear up-conversion of some longer wavelength driving field or the direct production of coherent light via free electrons either confined to a plasma or fourth generation accelerator based device.

II.A.3.1 Direct Frequency Conversion of Long Wavelength Lasers

The following assumptions are used to estimate the ultimate limits of creating ultra-high fields at short wavelengths by direct frequency conversion of ultra-high intensity lasers. Direct frequency conversion schemes are High Harmonic Generation, the VUV-emission of relativistic free electrons under the influence of an ultrahigh laser field, and Compton backscattering from high energy electrons.

II.A.3.1.1 HHG

The study of the interaction of atoms/molecules with intense laser fields has resulted in the understanding of the dynamics associated with the production of high harmonic radiation in gas jets. The refinement of experiment and theory have led the way for understanding not only the single atom response to the laser field but also the macroscopic propagation associated with the harmonic source. Current sources can produce coherent radiation out to 2 nm and a brightness exceeding third generation accelerator based light sources. The intensity produced by harmonic generation have thus far has been limited more by macroscopic target considerations then the fundamental bounds placed by the single atom-laser interaction.

If the technical constraints of the target can be overcome by some means of a quasi-phased matched extended source of atoms,ions and clusters then it is possible to estimate obtainable intensities. Assume a near visible drive laser of 1 PW peak power producing harmonic photons with a 1 keV limit and 10^{-6} power conversion efficiency. Using multilayer mirrors producing a spot size of 10^{-3} of the fundamental beam yields an intensity of 10^{22} W/cm^2, well in excess of an atomic unit of field. Furthermore, temporal

compression of the harmonic process could produce an additional factor of ten in the peak power, although the fundamental limit will be one optical cycle of the field or 20 as.

II.A.3.1.2 Free Electrons in Intense Laser Fields

Free electrons in intense laser fields undergo periodic ponderomotive oscillations. At intensities above 10^{18} W/cm^2 for visible-frequency lasers, this motion become relativistic. This has obvious importance for light-source generation, and has been explored in the context of interactions with relativistic laser beams and with electrons in plasmas.

The oscillating fields in lasers make them interesting candidates for high periodicity insertion devices in accelerators, and one could imagine such devices producing tailored vacuum-ultraviolet or x-ray radiation sources, possibly with high coherence.

In this context, we note several recent reseach projects in this area. (This is NOT a comprehensive list!)

Transverse Thompson scattering of photons from an ultrafast laser scattering from a linear accelerator electron bunch at the ALS has produced ultrafast pulses of hard x-rays.

A SLAC proposal would turn part of the linac into a FEL producing 1-10 angstrom x-rays.

SLAC has also produced 29 GeV gamma rays by Compton backscattering of an intense laser from the 46 GeV electron beam. Compton backscattering produces extremely intense, ultra-short radiation with associated high field strengths, which will be of main interest to the high energy physics community. While it is outside the scope of AMO applications, it represents one of the interdisciplinary links within the DOE program.

A wakefield acceleration experiment at Michigan has produced MeV electrons in sub-picosecond bunches, which could produce ultrafast x-rays via bremstrahlung.

II.A.3.2 VUV- and XUV-Sources and Amplifiers

An intriguing new vista of TW, short wavelength sources have been discussed in the context free electron lasers. The basic principle relies upon the efficient exchange between the photon field and relativistic electrons accelerated in the magnetic field of a wiggler. In this regard, two approaches are under investigation each with their own merit but based on a similar principle of high gain amplification. The SASE (self amplified spontaneous emission) scheme is based on the exponential amplification of radiation which builds up out of the noise in the first few sections of the wiggler. Thus, the technical limit of this emission will be based on electron beam energy and quality, e.g. emittance.

Both SLAC and the TESLA project at DESY have proposed operation at 1 nm with an intensity of 10^{20} W/cm^2. In the beginning of calendar year 1997, important proof-of-principle demonstrations of the SASE mechanism have been conducted at three major laboratories (Brookhaven, Los Alamos, UCLA) at infrared wavelengths. These results provide an important benchmark for testing the theoretical models which are being used to scale to shorter wavelengths. Current analysis have been promising.

A second approach (BNL and Jefferson Labs) is based on harmonic up-conversion of a laser-seeded FEL. The advantages of this scheme over SASE are in the coherent properities, peak power and pulse duration (as short as 3 fs) of the output light. However, the SASE scheme is able to produce light at shorter wavelengths (below 10 nm).

Based on recent current designs in the area of VUV-free electron lasers and X-ray lasers we estimate the following limits of intensity and field strength at wavelengths between 1 μm and 1nm:

@ 200nm (TJNAF FEL) design intensity 10^{17} W/cm^2
@ 100nm (BNL FEL) design intensity 10^{20} W/cm^2

@ 10nm (TTF FEL) design intensity 10^{17} W/cm^2
@ 1nm (TESLA FEL, SLAC FEL) design intensity 10^{20} W/cm^2
@ 1nm hypothetical x-ray laser (10 mJ / 1 ps) intensity 10^{24} W/cm^2

(Note that these are "back of the envelope" upper limits which don't take into account possible sub-optimal focusing characteristics in these sources.)

Conclusion: Current technology allows the assumption that the wavelength region between 1 μm and 1nm will be covered by various high-intensity (10^{20} W/cm^2), high-field (10^{11} V/cm) light sources, available to AMO physics.

II.A.4. Infrared Sources of High Intensities and High Field Strengths

While today's most intense solid state lasers operate in the near infrared around 1 μm, no materials or concepts are presently known to create coherent light at longer wavelengths (10 to 100 μm) with comparable intensities and field strengths. This is in remarkable contrast to the short wavelength region (1μm to 1 nm), where both frequency conversion methods and independent light sources exist at intensities around 10^{20} W/cm^2 and field strengths around 10^{11} V/cm.

From a physical point of view one expects lower intensities and field strengths for the following reasons:

a. The amount of energy stored in a given volume of an active laser medium of density rho decreases inversely with the wavelength of the radiation.
b. The maximum bandwidth (ultimately limited by the transition energy) also decreases inversely with the wavelength, thus lengthening the pulses and decreasing the maximum power.
c. The focal spot size increases proportional to the wavelength, thus decreasing the maximum intensity. Hence, for comparable volumes of the active medium the maximum intensity is expected to decrease at least with the fourth power of the wavelength.

II.B. Harnessing Quantum Dynamics

In this section, we would like to project a vision of harnessing quantum dynamics as an important enabling capability for the future. Here we have in mind not simply new probes of matter (be they optical or otherwise), but more generally, the ability to manipulate complex quantum systems to do our bidding with a degree of dexterity that is only glimpsed by current capabilities. Because the systems of interest are bound on energy scales given by electron volts over distance scales of Angstroms, the time scales implicit for these investigations are in the femto-second domain and beyond, and the intensities involved are greater than 100 terawatts per square centimeter. Hence, a necessary component of our vision is the need for the advancement of technical tools to a frontier well beyond current capabilities.

As pulsewidths have decreased and peak power and wavelength control has increased in the recent decade, it has become possible to conceive of a new regime of light-matter interaction in which the light is no longer treated as a passive observer. We are approaching an era in which we can "sculpt" dynamical systems to a large number of purposes, improving techniques for chemistry, fast microelectronics, optical communication, and even energy deposition in high density plasmas. Some tantalizing recent work pointing to this new area is the coherent control of Bragg scatterers at synchrotrons, which could provide a way to make high-brightness sub-picosecond x-ray sources (Larsson, Z. Chang, E. Judd, P.J. Schuck, R.W. Falcone, P.A. Heimann, H.A. Padmore, H.C. Kapteyn, P.H. Bucksbaum, M.M. Murnane, R.W. Lee , A. Machacek, J.S. Wark, X. Liu, B. Shan, Optics Letters 22 1012-1014 (1997)); production of wakefield plasmas for compact particle accelerators and ultrafast

sources of gamma rays and relativistic electrons (R. Wagner, S.Y. Chen, A. Maksimchuck, and D. Umstadter, PRL 78, 3125 (1997)); production of unique forms of radiation, such as "half-cycle pulses" (D. You, R.R. Jones, P.H. Bucksbaum, and D.R. Dykaar, Opt. Lett. 18, 290 (1993)); and tailoring of coherent xuv radiation from high harmonics (I.P. Christov, M.M. Murnane and H.C. Kapteyn, PRL 78, 1251 (1997)).

Each of these has important applications to basic science and to energy research; each also presents special opportunities for contact to other areas of science and technology.

II.B.2. Detailed Manipulation of Quantum Systems

The tools are now at hand that one can begin to realize the long-sought goal of reaching inside of a quantum system and manipulating it with the same ease that one can manipulate a macroscopic system. Of course, the term "quantum systems" encompasses an enormous range of topics, and there is no one set of tools and methods that will allow manipulation of all of them. At present there are some particular systems that are promising for achieving this detailed level of control.

Among these are the control of the center-of-mass degree of freedom of atoms and ions in traps; the control of the vibrational degree of freedom in a molecule, and the internal electronic degree of freedom in an atom.

The most important applications that will be available if this capability is realized are no doubt yet to be conceived, but some are obvious: the control of chemical reactions, the production of materials with unique optical and electrical properties, and the development of better understanding of quantum dynamics. There are less obvious possibilities of some promise including massively parallel computing, and secure communications.

II.B.3. Pulse shaping and adaptive control

II.B.3.1. What and Why

Behavior of quantum systems is determined by the time-dependent Hamiltonian and initial conditions. Therefore, modifications of this behavior are made possible by the application of external fields that change the Hamiltonian and/or the initial state of the system. Developments in optical technology have now made it possible to envision detailed manipulation of quantum systems using carefully shaped electromagnetic pulses. This capability will allow the development of technologies at an entirely new sub-atomic scale, and at the same time allow the utilization of uniquely quantum mechanical properties for practical application. There are classical analogies to certain types of manipulation. For example, fields might push an electron in a certain direction within an atom, deposit energy in a specific chemical bond, or induce directed electron currents in solids. However, there is also an inherent complexity in these systems due to their quantum nature. While this complexity can make simple classical manipulation more difficult or impossible, it also provides additional degrees of freedom for non-classical control. For example, states of multi-electron atoms and molecules are naturally correlated and entangled. These uniquely quantum features are exactly what is needed for applications in quantum computing, secure encryption, and other areas not yet envisioned that rely on the ability of quantum systems to explore simultaneously many parallel paths.

II.B.3.2. What do we need?

To realize arbitrary manipulation, tools are needed to shape or mold the quantum wavefunction. These tools are electromagnetic fields and the interaction Hamiltonian. Wavefunction "sculpting" has two basic requirements. First, the amplitude of the electromagnetic field must have temporal variations over periods comparable to the characteristic dynamical time-scales of the system. Second, the field amplitude must be sufficient to cause a significant change of the quantum mechanical state. Therefore,

the development of sources that can provide intense, coherent, broad-band radiation is a prerequisite to quantum manipulation. Furthermore, optimal control cannot be achieved without the ability to arbitrarily alter the time- dependent structure of the fields that are used to sculpt the quantum state. In a very real sense, the prospects for quantum manipulation are tied directly to our ability to alter coherent electromagnetic fields at will. In turn, exquisite field manipulation requires currently unrealized control of the properties of optical materials.

II.B.3.3. Problems to be solved

Incremental progress is currently being made in manipulating dynamics in atomic, molecular, and condensed matter systems. However, the ultimate control described above cannot be realized until solutions to extremely challenging problems have been found. First, new laser and electro- optic materials must be developed to facilitate the production and control of the requisite fields. The new frontiers for increasing the coherent bandwidth, frequency, and intensity of laser sources is discussed in Section III A above.

Second, in most systems of interest, the complexity of the Hamiltonian and number of degrees of freedom in the system prohibits the a priori determination of the most appropriate time-dependent field. Therefore, active feedback techniques must be developed to adjust the field characteristics until the final quantum state converges to the desired configuration. Implementation of adaptive feedback requires that several obstacles be overcome. First, methods for detailed control of the time-dependent fields must be developed. This is no small task considering that in some situations sub-femtosecond temporal structure will be required. Second, experimental methods for determining the time-dependent quantum state of the system must be discovered and implemented. Such techniques will require high spatial and temporal resolution in each dimensional coordinate and will necessarily rely on the generation of tailored light pulses. Third, the efficient algorithms that are currently used in classical control problems must be adapted to specific multi-dimensional quantum problems. These algorithms will rapidly interpret the quantum state feedback and produce an error signal for input into the field shaping device.

II.B.3.4. (Im)Possible Dreams

In the following subsections, we outline potential applications of quantum manipulation to problems of fundamental scientific, and possibly practical, concern. We do not imply that this is an exhaustive list. Because the realization of these dreams is inexorably linked to future technological developments, we cannot provide specific example systems or experimental details.

II.B.3.4.1. Controlling Coherent Collisions

At first glance, the random nature of collison processes seems at odds with the concept of a coherent collision. However, there does not seem to be any fundamental physical restriction which would prevent the use of coherent radiation to affect collision dynamics. For example, tailored light pulses could be used to enhance or inhibit particular photo-assisted reactions at specific times. Alternatively, coherent fields could be used to produce an initial atomic or molecular state which is transparent to collisions at specific energies and/or the relative orientation of colliding particles. Conversely, the atomic or molecular wavefunction could be manipulated so that it reacts at specific times, and only in certain reaction coordinates. Lastly, dynamics established in the internal degrees of freedom of an atom or molecule, A, might be transferred to a collision partner, B. In this case the collision dynamics would facilitate excitation of internal direct application of radiation fields.

II.B.3.4.2. Quantum Encoding

Secure communication is an important and ongoing problem. The properties of quantum systems make them potential useful for encryption applications. For example, information (phase and amplitude) can be encoded into the wavefunction of a complicated

quantum system with many entangled degrees of freedom. After a very short time, phase evolution makes the code unrecognizable. A time-dependent field applied to the system at any time after the creation of the coded state acts as a "key" which produces an echo of the initial state. The key can have limited access and be universal so that it is capable of retrieving any code stored in that system.

II.B.3.4.3. Single Atom/Molecule Electronics

Tailored light pulses produce a coherent quantum state in an atom/molecule/nanostructure. This quantum state acts as a transistor, switch, or logic gate. Digital data or analog signals are input to the device using an additional electromagnetic pulse. A third pulse interogates the quantum state of the device providing the readout. The complexity of device function is determined by the complexity of the initial configuration field as well as the I/O pulses. In fact, single function quantum logic gates have already been produced using trapped ions

II.B.3.4.4. Bond-Selective Chemistry Through Coherent Excitation

There has been recent progress towards this long-discussed goal of physical photochemistry. Multiple interfering pathways toward specific ionization or dissociation products has been demonstrated (L. Zhu, et al., Science 270, 77 (1995)). Ultrafast coherent wavepacket control in molecules has also been observed (B. Kohler, et al., PRL 74, 3360 (1995)). Since most interesting photochemistry involves systems in contact with rapidly dephasing media such as solvents, a great premium is placed on ultrafast, coherent, and strong fields that can perform their quantum-mechanical tasks in only a few tens of femtoseconds, before dephasing of the system. Furthermore, since the Hamiltonian for these systems can be quite complicated, and only known approximately, adaptive control strategies will be quite important.

Coherent control in the strong field limit may solve two fundamental problems in bond selective chemistry: dephasing, and low yield. This is because strong fields can dominate the energy scale in the problem as they reshape the molecular system. For recent work in this area, see G.N. Gibson, M. Li, C., Guo, and J. Niera, Phys. Rev. Lett. 79, 2022-2025 (1997), and E. Constant, H. Stapelfeldt, and P.B. Corkum, Phys. Rev. Lett. 76, 4140-4143 (1996).

II.B.4. High Intensity Optical Channeling.

When high intensity light propagates through a transparent medium, its presence alters the optical properties of the medium. The resulting self phase-modulation and self-focusing have already found many applications in short pulse laser technology, micromachining and optical propagation. Other similar phenomena are electromagneticaly induced transparency and "photon bullets," which are stable conducting plasma channels. The latter have been suggested for remote lightning protection.

As the power scales up, the nonlinearity changes in some important ways. Above a critical whole-beam power threshold, the principal nonlinear self-focusing mechanism becomes relativistic self-focusing. The consequences of this are pretty startling: An intense laser beam focused into a plasma in this regime forms a cavitation channel, a region of low or even zero electron density, in which the laser can penetrate for long distances. The channel formed in this way can be tailored by shaping the intense optical pulse to optimise a number of physical properties, such as the density, width, length, or field gradients.

One important application of this technology is transmission of the bulk of the laser energy deep into a plasma, where it can be deposited at a plasma density gradient. Such schemes may be critical for efficient energy conversion using laser-compressed fusion targets (ref: Fast Ignitor workshop).

As important as this is for future energy development, it is only a small part of the potential applications of

high intensity laser-channeling. This expulsion of electrons coupled to relativistic self-focusing then produces a large (GeV/m) longitudinal wake field which can accelerate bunches of electrons to multi-MeV energies over millimeter distances. Such beams have already been shown to have some unique properties, such as short bunch length and narrow divergence.

Applications to the needs of traditional particle acceleration are obvious: the tremendous increase of the field gradient in these plasmas can lead to great scaling advantages for small accelerators. There may be some unique opportunities here as well to take advantage of the novel conditions in the plasmas. For example, the bremmstrahlung radiation produced when these ultrashort electron bunches hit a solid target may provide the best short pulse technology for ultrafast gamma radiation.

II.C. Matter in Extreme Conditions

Advanced laser and particle sources providing short-pulse and high-intensity photon, electron, and ion beams are expected to become primary enablers of AMOP science and technology in both the near and distant future. Each source has spatial, temporal and spectral characteristics that are unique and complimentary. They will allow controlled deposition of energy into various systems, producing states previously inaccessible in earth bound laboratories. They will permit studies of the atomic physics of matter under extreme and unusual conditions, which include: extreme pressure (Gbar) extreme fields (E >> 1 keV/Angstrom, B > 1 gigagauss)

II.C.1. Extreme Pressure

Everything from Fermi-degenerate gases, nonequilibrium plasmas and highly perturbed atomic systems can be studied in these interactions. Extreme pressures are found in astrophysical objects, pulsed power machines, weapons, advanced x-ray sources, and ICF fusion pellets. They may also enable new routes to IC fusion such as the fast ignitor. The transition of the atomic structure of matter from its different states (eg. from solid to liquid to gaseous phase can be observed with unprecedented temporal resolution. Matter can also be heated isochorically (without change in density) to extreme temperatures (kilovolts). These studies are important for the understanding of radiation damage, material ablation for thin-film deposition, fusion, pulsed power, propulsion, waste disposal and plasma processing. The x-ray emission from the laser-heated solid-density plasma also makes a bright, ultrafast and coherent table-top x-ray source that can be used to study transient phenomena on an ultrashort timescale. Such a source can also be used to study nonlinear optics with inner-shell transtions. It is relevant to x-ray lithography and indirect drive ICF. In the latter case, the source can be used as an imager to freeze hydrodynamic motion and provide a test bed for the study of material properties at a wavelength relevant to hohlraums.

II.C.2. Extreme Fields

When the work done on an electron by the field over the distance of the laser wavelength equals the electron rest mass (10^{18} W/cm^2), we enter a new regime of nonlinear optics. In this case, a plasma medium can be modified by the laser field in such a way that it can be used to guide the laser light at high intensity over a much greater distance than it would in vacuum. The light pipe that results when the electrons and ions are driven from the axis of the channel can be used to guide another laser pulse. This is the basis for the proposed fast-ignitor fusion concept. Gigagauss magnetic fields and electron-positron plasmas may be created. Interesting ionic states may be created and relativistic effects dominate the interaction physics. The largest electrostatic fields ever produced in the laboratory are generated in wakefield plasma waves, which can accelerate useful numbers of electrons to gigavolt energies in a distance of just a centimeter. This is a new regime of parameter space that may result in new properties of matter and is relevent to advanced accelerators, electron sources and fusion energy concepts. One of

the exciting possibilities opened by high-intensity laser and ultra short relativistic electron and ion beams is the creation in the laboratory of extremely high electric and magnetic fields at the critical Compton limit of 10^{16} V/cm. In these extreme fields, the relation between energy and the vacuum are modified in a dramatic way. One expected consequence is the breakdown of the vacuum. In addition, the large energies deposited over a macroscopic volume would be sufficient to generate collective and coherent effects. This will result in the formation of an e+e- plasma. Just as one needs a theory of non-linear optics to understand strong electromagnetic interactions, a theory of non-linear QED will be needed to understand these collective effects.One concept for achieving these fields coherently backscatters an intense femtosecond laser from relativistic electron beams to generate fields on the order of 10^{29} W/cm^2. Coherent effects in the backscattering would then enhance the field by ne^2.Also, laser-based sources can be built for the generation of energetic electrons (laser accelerators), ions or neutrons. The pulse duration of these electron bunches can potentially be subfemtosecond, which is sufficiently short to lead to novel effects. For instance, coherence effects can enhance the intensities when the electrons in the beam are acting collectively (scaling as N^2 instead of N). The duration can also become shorter than several interesting characteristic timescales, such as Auger lifetimes.

II.C.3. Connections to Other Fields at DOE

High field AMO physics experiments are related to the nuclear physics studies of interactions between relativistic heavy ions at RHIC. In contrast to the interactions at RHIC, which involve quarks and gluons, the AMO experiments described here involve photons and leptons. Studies of the interaction and creation of particles in these experiments may be a subset of those observed at RHIC. Because there are fewer types of interactions interpreting these experiments maybe be less complex. In addition, the intense fields found in pulsars may be recreated by high power laser lasers (or other sources). In these intense fields, atoms may undergo recombination with electrons or other particles that are pulled out of the vacuum.

Panel C: Surface Interactions With Photons, Electrons, Ions, Atoms, and Molecules

Chair:
J. Wayne Rabalais (University of Houston)

Panelists:
Raul A. Baragiola (University of Virginia)
Barbara H. Cooper (Cornell University)
F. Barry Dunning (Rice University)
Charles S. Fadley (Lawrence Berkeley National Laboratory)
J. William Gadzuk (National Institute of Standards and Technology)
Wilson Ho (Cornell University)
Dennis C. Jacobs (University of Notre Dame)
Theodore E. Madey (Rutgers University)
Steven J. Sibener (University of Chicago)
Nicholas Winograd (Penn State University)

Executive Summary

NEEDS: The present understanding of the interactions and behavior of particles (ions, atoms, molecules, clusters, electrons) and photons with surfaces at the basic atomistic level is far from complete. A large number of outstanding questions remain that pertain to, for example, the detailed dynamics of particle-surface interactions, localized energy deposition at surfaces, nonequilibrium phenomena, and the properties of atoms and molecules on a surface. Central to improving the present understanding is control of the energetic, spatial, and temporal degrees of freedom of the particles involved in the interactions, coupled with manipulation of atoms and molecules at the surfaces. Improved understanding of surface phenomena is fundamental to the development and synthesis of materials with specific physical and chemical properties. This underpins future technological advances in areas such as energy production and conservation, microelectronics, and the environmental and life sciences. Many of these applications are based on phenomena that occur at surfaces and interfaces, e.g. solid/vacuum, solid/gas, liquid/vapor, and solid/liquid, as a result of stimulation by particles or photons. Studying such phenomena has historically been difficult and challenging. Although much progress has been made to date, the microscopic understanding of many interfacial phenomena remains below that currently existing for homogeneous phases, i.e. gas-phase atomic and molecular physics and solid state physics.

OPPORTUNITIES: A unique opportunity currently exists to advance this challenging area of basic science. This involves the integration of the expertise and technology from atomic, molecular, and optical physics (AMOP) with that of surface science. AMOP scientists have developed an in-depth understanding of the properties and behavior of gas phase atoms and molecules, highly sophisticated experimental techniques to produce and analyze particle beams in well controlled states, and advanced theoretical methods. Surface scientists have developed a comparable battery of techniques for control of the state of a surface and improved theories, particularly for thermal processes at the gas/surface interface. The relatively recent advances in the production of large clusters of atoms and molecules represents an area that is naturally intermediate between AMOP and surface science. Each area has much to learn from the expertise of the other. For example, for almost every particle-surface interaction, there exists a comparable analog in gasphase collisions.

BENEFITS: This synergistic relationship between AMO and surface scientists will enable new advances in:

- experiments that involve selection, control, non-thermal chemical manipulation, and complete analysis at the atomic and molecular level.
- theory that couples the discrete excitations of atoms and molecules with the continuum excitations in solids.
- the use of molecules and clusters for achieving an atomic level understanding of the mechanisms responsible for chemical catalysis and gas-surface energy transfer.
- novel schemes for materials growth and nanostructure fabrications.

The synergy will facilitate progress in the important areas of: (1) dynamics of excited states at surfaces, (2) non-equilibrium phenomena induced by particles and photons at surfaces, (3) control and manipulation of particle- and photon-surface interactions at the atomic and molecular level, and (4) advanced techniques for probing particle- and photon-surface interactions.

Outline

I. Fundamental Understanding of Particle- and Photon-Interactions with Surfaces
II. Non-Equilibrium Phenomena
  A. Low Density Excitations
    A1. Non-Equilibrium Conditions in Film Growth and Modification
    A2. Theoretical Approach
    A3. Technological Importance
  B. High Density Excitations
    B1. Dynamics of Electron Transfer
    B2. Dynamics of Highly Concentrated Excitations
    B3. Mechanism of Energy Flow in Collisions
    B4. Particle-Surface Interactions as Probes of Surface Dynamic Properties
    B5. Laser Induced Non-Linear Phenomena

III. Control and Manipulation at the Atomic and Molecular Level
  A. Energetic Control and State Selection
  B. Spatial Control
  C. Temporal Control

IV. Other Advanced Techniques for Probing Particle- and Photon-Surface Interactions
  A. Beams of Ions, Atoms, Molecules, Clusters, Electrons, and Photons
  B. Advanced Combinations of Supersonic Beams, Atomic Traps, and Atom Optics
  C. Femtosecond (fs) Surface Spectroscopy
  D. Spatial- and Temporal-Imaging of Surface Atomic Structures
  E. Coincidence Techniques
  F. Surface Studies in High Pressure Environments
  G. Higher-Efficiency and Higher-Speed Detectors

V. Key Research Areas in this Report that are Under-Represented in the United States

VI. Summary List of Basic Unsolved Problems Involving the Interactions of Low Energy Particles (Electrons, Ions, Atoms, Molecules, Clusters) and Electrons with Surfaces

VII. Summary List of Applications of Particle and Photon Interactions at Surfaces

VIII. Conclusion Statement

I. Fundamental Understanding of Particle and Photon Interactions with Surfaces

Considerable progress has been made over the past several years in describing the properties of clean and adsorbate-covered surfaces. Less is understood about the dynamical processes that occur when particles (electrons, ions, atoms, molecules, and clusters) and photons impinge on a surface, especially when excited states are involved. A detailed knowledge of the formation and relaxation of excited states is of principal importance in understanding nonequilibrium processes and in exploiting these processes in novel applications.

Excited electronic states are formed near the surface as a result of the absorption and scattering of photons and electrons and by the impacts of ions, atoms, molecules, and clusters. Energy is also deposited directly into nuclear motion by momentum transfer to surface atoms. These impact-induced excitations range from, for example a change in the charge state of a low-energy scattered projectile or an electron-hole pair creation by the absorption of a photon, to high density excitations such as track formation and material ablation resulting from heavy or highly charged ion impact. State-of-the-art experiments utilizing surfaces that are well-characterized, i.e. atomically clean, aligned structures, and highly controlled beams of particles that are mass-, energy-, charge-, and state-selected, collimated, and pulsed (in some cases), in conjunction with techniques for complete delineation of the interactions in terms of elemental-, spatial-,

temporal-, and final state-resolution, are needed in order to reach a new level of fundamental understanding.

Beam-surface experiments that probe the fate of a scattered projectile or that follow the nascent excitations in the surface are required to provide a comprehensive understanding of the relevant energy and charge exchange mechanisms and the relaxation of excited states. For example, final electronic state distributions of scattered atoms or molecules, coupled with theoretical models of nonadiabatic electronic interactions, reveal the charge transfer mechanisms and information about the lifetimes of excited states near surfaces. Desorption of atoms or molecules following electron impact indicates the coupling of nuclear motion to electronic excitations. Ultra-low and thermal collisions of neutral atoms and molecules with interfaces are also of high interest, with such gentle interactions dominating phonon exchange, sticking, and chemical reactivity. Fragmentation patterns following cluster impacts reflect energy redistribution pathways and the time scales over which energy redistribution occurs. Electron emission subsequent to impact identifies the specific excited states produced in the projectile and/or substrate. Moreover, time-resolved spectroscopies involving ultrafast lasers or third generation photon sources will allow one to directly follow how electronic excitations couple to other degrees of freedom and how localized excitations decay in space and time. Experimental advances, focused on increasing control of the incident species and improving temporal and spatial resolution for probing the final states, will provide new windows for understanding dynamical pathways.

New theoretical developments are essential to accurately treat the inherent many-body nature of particle-surface interactions, including electronically excited states at surfaces and relaxation pathways. One of the challenges is the treatment of phenomena that evolve over a time scale of some ten orders of magnitude. A simplistic delineation is as follows: (1) The collision regime (< 100 fs) involving direct nuclear collisions, excitation of electronic transitions, creation of a local non-equilibrium, high-energy-content region (mini-collision cascade), and generation of vacancy and interstitial pairs. (2) The thermalization regime (0.1 - 10 ps) involving dissipation of excess energy through phonons, rapid quenching of the cascade region, and material recrystalization. (3) The diffusion and recombination regime (1 ns - 10 μs) involving thermal diffusion and annihilation of point defects by diffusion to recombination sinks. Meeting this theoretical challenge requires well coordinated expertise from many areas in AMOP as well as from chemical and condensed matter physics. Theoretical modeling and simulation of the time evolution of an excited atomic or molecular system coupled to a surface (with all its unique attributes), possibly in an intense AC field (laser) or DC field (scanning tunneling microscope (STM), tunnel junction, field ion or emission tip), requires creative new approaches transcending brute force diagonalization of large matrices or numerical solution of large but finite systems of ordinary differential equations which incorporate standard force laws or manageable potential energy surfaces.

Because of the importance of electron charge redistribution and transfer, quasi-localised resonance state formation, and field-induced electron dynamics, the coupled, multi-dimensional quantum equations of motion must be able to treat the electronic and nuclear dynamics on an equal footing. An additional challenge arises because the solid and laser fields each present a continuum of excited states that must be considered and exploited within theoretical models. Low and ultralow energy interactions present another exciting challenge due to the unusually large deBroglie wavelengths and sharp resonances which characterize such interactions. Surface charge density distributions and collective surface phonon modes must receive accurate treatment if such calculations are to realistically model atomic scale behavior. It is envisioned that innovative combinations of appropriately statistically averaged classical (Newton, Langevin equations, etc.), semiclassical (WKB,

Gaussian wavepacket dynamics, etc.), and quantum (coupled channels, wavepacket propagation on grids, soluble model Hamiltonians, etc.) particle-field models will be required, together with intuition for differentiating the relevant from the irrelevant aspects of the interacting system. This work should provide a tractable, useful, and informative theory on fundamental processes at surfaces that can serve to guide, interpret, and predict experimental results. Close experimental-theoretical coupling must be maintained, each helping to guide the other in this "self-correcting" drive to achieve a fundamental understanding of the interactions involved. This, in turn, will lead to unprecedented control and exploitation of these interactions in important applications such as nanoscale surface modification, low temperature growth of novel materials, and creation of metastable phases.

II. Non-Equilibrium Phenomena

Energetic particle and photon beams provide a means of producing conditions in the surface region that are far from thermodynamic equilibrium. Understanding the fundamental particle- and photon-surface interactions will improve our ability to exploit them. The new degrees of freedom resulting from the use of controlled beams allow non-equilibrium excitation of solids, which can be used to deposit films, synthesize new metastable materials and phases and understand radiation effects. This section is roughly divided into two subsections, i.e. low density and high density excitations.

A. Low Density Excitations

By low density excitations we refer to those created in the near surface region by, for example, the impacts of relatively low energy atoms, molecules, and ions.

A1. Non-Equilibrium Conditions in Film Growth and Modification

Particles of a wide range of incident kinetic energies can selectively influence film growth chemistry depending on the nature of the potential energy surfaces which govern such processes. Consider, for illustrative purposes, the energy regime that is directly applicable to film growth and modification on surfaces involving appreciable entrance channel barriers. In this instance, a reasonable lower limit is a kinetic energy of the order of bond energies, *ca* 1 eV. The energy released upon bond formation is dissipated as phonon-like excitations and lattice distortions which perturb the local environment in a manner similar to the excess kinetic energy supplied by the incident beam. At this lower limit, chemical bonding interactions become significant, the binary-collision approximation becomes questionable, and inelastic interactions between the incident particle and substrate can alter the incident trajectories. A reasonable higher limit, in the case of ions, is a kinetic energy of the order of a few keV. In this range the sputtering yield becomes equivalent to, or higher than, the beam flux, classical ion-trajectory simulations using a binary-collision approximation provide a satisfactory description of many of the collision phenomena, and the impinging ions remain in the near-surface layers. The thresholds for penetration, displacement, sputtering, and reaction occur in this energy range. In the case of neutral atoms, supersonic beams can easily span the range from sub-thermal energies up to several eV. This allows one to selectively and systematically examine chemical reaction channels which become accessible at ever increasing energies. Some of these channels may lead to new or more highly optimized growth conditions than are typically accessible in CVD environments. Moreover, supersonic beam experiments permit intentionally selected growth precursors to impact the target sample, greatly simplifying the analysis of growth mechanisms. The ability to select the energy, type, and arrival rate of particles, incident polar and azimuthal angles, substrate temperature, background gases, and stoichiometry allows control of the penetration and defect energy thresholds, film growth mechanisms, isotopic composition, low temperature epitaxial growth, chemical reaction mechanisms, collision-induced surface chemistry, energy flow in collisions of molecular species, and coupling of gas and solid phase

excitations. While conventional film growth techniques rely on high temperature to activate chemical reactions and diffusion processes, growth of novel high quality materials at low temperatures can be achieved by coupling the internal and translational energy of the incident particles with the surface energy modes. Low temperature growth is becoming increasingly important in a number of technological applications where it is desirable to prevent interdiffusion at interfaces and to stabilize nanoscale structures. The fundamental mechanisms distinguishing energetic beam deposition from thermal techniques result from localized deposition of energy by the incident particles. The energy stored in the incident particles can be used to overcome reaction barriers and induce nonthermal chemistry. Electronically excited states of impinging atoms is an example that is open for exploration. Nonequilibrium surface atom configurations can change nucleation and growth modes, which include processes such as adatom and vacancy production and enhanced interlayer mobility. The challenge is to learn to balance the beneficial effects of ion irradiation, such as local relaxation, creation of mobile vacancy/interstitial pairs, and enhanced diffusion, with the undesirable effects, such as permanent defect formation, lattice damage, sputtering, atomic mixing, etc. It is necessary to understand the role of particle energy, mass, and charge in penetration of surfaces, displacement of atoms, stimulation of chemical reactions, production of metastable structures and high density materials, and enhancement of epitaxy.

Other non-thermal processes in surface modification by energetic beams include electron- and photon-stimulated desorption and reactions. These processes involve electronic excitations that lead to bond-breaking. The manifold of possible electronic excitations allowed in gaseous molecules may be very different in the condensed phase. Many challenges remain in understanding energy transfer, charge transfer, and the dynamics of excited-state evolution. An improved understanding of energy flow in thermal energy collisions is also needed if we are to predict energy transfer between impinging neutral particles and clean/adsorbate covered surfaces. Such interactions, typically mediated by phonons, are crucial to an improved understanding of surface phonon spectroscopy, i.e. bonding at surfaces, sticking, and energy accommodation in general.

A2. Theoretical Approach

A full understanding of the interactions involved will require the more sophisticated atomistic simulations that are currently emerging. A central problem that needs to be resolved is the importance of the coupling between electrons in the solid and the moving atoms in the collision cascade initiated by the energetic interactions. Molecular dynamics simulations of radiation effects have incorporated this special case of electron-phonon coupling in a parametric form, and show that solidification and phase transformations in the energetic atomic cascades are affected by the uncertainty in this coupling. Recent attempts have focused on ab-initio descriptions of energy levels and energy transfer in energetic cascades, but this type of theory is still in its infancy. Improvements are needed in calculating the fate of reactive molecular precursors with reactive interfaces. Sophisticated electronic structure calculations analogous to those developed for quantum chemistry and precision atomic physics applications would have high impact in this area of endeavor.

A3. Technological Importance

Many techniques employ energetic particles in the growth process. These include sputter and plasma deposition, pulsed laser deposition, and ion-assisted and direct ion beam deposition. These techniques often involve a wide range of incident species and energies. Experiments with highly controlled incident beams (ions, neutral molecules, clusters, photons) and real-time *in situ* diagnostics are critically needed to gain a better understanding of the role of energy in modifying growth modes and materials properties. This new understanding will enable us to control important film properties such as structure, stoichiometry, internal strain, surface and interface roughness, and adhesion. Energetic beams have been

shown to have beneficial effects in the growth of magnetic multilayer materials and wide band gap semiconductors (SiC, Group III nitrides, and diamond). This approach shows great promise for controlled growth of novel materials and metastable phases. In order to achieve spatially resolved deposition and surface modification, it is desirable to develop new techniques relying on atomic optics and ion beam focusing. Of course, energetic electron and photon beams will continue to be of critical importance for lithography in microelectronics applications as dimensions are reduced to the sub 100 nm range.

B. High Density Excitations

An exciting new development in the field of interactions of particles and photons with surfaces is to produce extended regions in solids that are excited far from equilibrium conditions through the impacts of energetic heavy ions and slow highly charged ions, clusters or large molecules. The highly unusual conditions which result cannot be described with existing models of particle-solid interactions. Questions of interest include the following:

B1. Dynamics of Electron Transfer

Interactions between ions and surfaces present new challenges which arise from the many-body target and its low geometric symmetry. Well-defined experiments involving ion neutralization and charge exchange at surfaces are needed to probe thresholds for excitation and ionization, to examine energy level shifts and broadening, and their dependence on particle velocity, surface electronic structure, and crystal structure and to investigate the effects of external fields and hybridization of electronic states near surfaces. The use of highly charged ions (HCI) provides a test bed for models that go beyond the independent particle description. HCI can range from He^{2+} to U^{92+} . At the upper end of this charge scale, they are a source of a wide variety of new physics. The electric field produced by a HCI and its image can induce enormous distortions of the surface potential of a solid, stimulating multiple electron transfers that result in the transient formation of “hollow atoms”. Electrons evolve in unusually high *n* orbits which would resemble Rydberg states except that they are polarized by the huge fields and they merge into extended electron states of the solid.

B2. Dynamics of Highly Concentrated Excitations

Ionization spikes (dense plasma) in non-metals, their evolution in some cases to a dense exciton gas, and their role in particle ejection, and non-thermal transformations in the solid (cratering, bond-breaking and reforming, phase transitions, etc.) cannot be explained by existing theories. These phenomena, that can be started by fast ionizing particles or slow highly charged ions, are important radiation effects in living tissue and semiconductor devices. A frontier in this area is the development of models for the complex, multiple-particle excited region and the time-varying, inhomogeneous electric fields. This can be accomplished by a new generation of molecular dynamics simulations which can include the appropriate local electric fields and many body interactions between ions, excited atoms and molecules.

B3. Mechanism of Energy Flow in Collisions

The flow of energy in collisions of clusters and large biomolecules with surfaces is a complicated, albeit very important problem. Current atomistic and continuum mechanics approaches are found, particularly for large, fast projectiles, to be insufficient to explain the coupling and evolution of vibrational and electronic excitations in the cluster and at the surface. These interactions result in extreme phenomena that include the formation of very hot plasmas in the solid, accompanied by photon and particle emission. This can lead to shock waves propagating through the material, cratering, and ejection of material. The projectile cluster can disintegrate in the collision or, at low velocities, be reflected in an unusual excited state, which includes

shape distortions, internal shocks, and vibrations. The relaxation of the cluster can result in fragmentation and thermionic electron emission and evaporation. The latter permits study of highly localized thermal phenomena. A goal here is to obtain complete characterization in terms of the underlying mechanisms, including molecular and surface quantum state-specific excitations, reactive processes, molecular fragmentation, and dissociative deposition. Controlling the many reaction channels is also of interest, as are interactions with complex surfaces (frozen gases, organics, polymers, clusters).

B4. Particle-Surface Interactions as Probes of Surface Dynamic Properties

Plasmons, collective excitations of valence electrons, provide an example of electron correlation effects that can occur at surfaces or in the bulk of solids and small particles. A long standing fundamental problem in solid-state physics has been how plasmons form and evolve in time and space, including their decay into quasiparticle excitations. It may soon be possible to study this important problem as a result of the development of new ultrafast laser techniques that provide a time resolution smaller than, or of the order of, the response time of the valence electrons (10^{-16} - 10^{-14} s). Surface plasmon excitations by photons and high energy particles can be monitored optically for real-time characterization of surfaces during materials processing and synthesis.

B5. Laser Induced Non-Linear Phenomena

Non-linear phenomena in the coupling of high power lasers with solids can lead to ejection of material from the surface. These phenomena induce a wide range of physical and chemical processes, depending on the excitation density, that have analogies and differences to those caused by ions. The unusual states of matter that are produced can be studied using AMOP techniques, such as picosecond (ps) lasers, sub-ns particle detectors, and coincidence techniques. High power lasers can be used for materials synthesis, micromachining, desorption, and other surface modification processes.

III. Control and Manipulation at the Atomic and Molecular Level

The previous two sections have focused on experimental and theoretical approaches to address unanswered questions regarding fundamental particle- and photon-surface interactions, and the non-equilibrium excitations that they entail. We have stated that an increased understanding of these interactions will enhance our ability to exploit them. Here we elaborate on techniques and approaches that will ultimately lead to our ability to understand, control, and manipulate these interactions at the atomic and molecular level.

Understanding of the physical and chemical properties of materials and of the means for achieving energetic-, spatial-, and temporal-control and manipulation of atoms and molecules are the fundamental underpinnings upon which nanotechnology is based. Atoms and molecules impinging on a surface can now be prepared in specific translational, electronic (exited ions and neutrals), spin, and internal states (vibrational and rotational), providing well-defined reagents. Once adsorbed, selective and nonthermal chemical processes can occur, driving a diverse body of research. The scanning tunneling microscope (STM) has become a powerful tool with the ability to probe and manipulate single atoms and molecules. Using femtosecond (fs) lasers, it is possible to access electronically excited intermediates and tailor intramolecular response dynamics. Both the tunneling electrons from the STM and photons from fs lasers can induce forces associated with select electronic excited states and can be used to “push” constituent atoms along desired reaction pathways, including motion on the surface, the breaking and forming of individual bonds, and the desorption of products. From measurement of the final translational and internal state distributions of the desorbed molecules, further insights into the mechanisms driving atomic and molecular motion can be obtained. Since charge transfer underlies many of the mechanisms involved in atomic and molecular manipulation and reaction, it is important to study electron interactions with atoms, molecules, and

clusters at surfaces. The energy levels and spatial distribution of excited states of atoms and molecules are directly involved in these interactions. Little is known about these states and theoretical investigations are critically needed for guiding the interpretation of experimental results and in making predictions of how individual bonds are made and broken. For electron and photon interactions with surfaces, the goal is to understand the mechanisms of excitation, ionization, electron capture, desorption, sputtering, reactions, damage, ablation, and charging of insulators.

A. Energetic Control and State Selection

Materials modifications can be controlled by impacting the surface with atoms and molecules prepared in specific energetic states. High translational energies can be used to overcome reaction barriers. Molecules in one vibrational state may be more effective than another in breaking or forming a chemical bond. Electronically excited atoms and molecules can also promote chemical reactions, as is evident in plasma assisted deposition and etching. By preparing atoms and molecules in well defined excited states, it is possible to unravel the mechanisms and dynamics of many complicated reactions in catalysis, atmospheric chemistry, and plasma reactors. State selection of atoms and molecules offers another dimension in the control of materials and the opportunity to tap into the extensive tools which have been developed by the AMOP community. In addition to obtaining a basic understanding of fundamental interactions between atoms and molecules with surfaces, state selection is expected to result in reactions which cannot be accessed by atoms and molecules with a thermal distribution. For example, new adsorption sites can be occupied and reactions can occur on entirely different parts of the potential energy surface. Species on the surface resulting from such reaction pathways can be probed by the STM. By creating an interference pattern on the surface with photon and atom beams, nanoscale structures can be formed on the surface which are composed of materials with novel structures and compositions.

B. Spatial Control

Recent advances in scanning probe technology make it possible to study single atoms and molecules, providing an unprecedented opportunity to understand and control materials. It is now possible not only to image with atomic resolution, but to obtain spectroscopic information and to induce chemical reactions with single bond precision. By examining individual atoms and molecules, it is possible to isolate environmental effects and obtain their intrinsic properties. Results from such studies are particularly attractive to theoretical investigations. Since tunneling electrons are confined to atomic dimensions, manipulation of single atoms and molecules is possible, providing the opportunity of building materials from basic atomic units. Atoms and molecules can be arranged into specific configurations, leading to nano-materials with novel properties. The ability to control and manipulate individual atoms and molecules and to induce chemical reactions with atomic resolution is the basis for single electron devices with ultra-small dimensions which can operate at room temperature.

Another exciting and AMOP-based opportunity involves stimulation of new research activities in gas-surface atom optics, incident beam control, and pattern formation. The use of either physical, electromagnetic, or optical fields can lead to well-focused beam spots of selected reagents on surfaces. Standing wave-interference patterns, already in use for such purposes, are but one application that will come from this new field of endeavor. This may well lead to the controlled fabrication of nanoscale structures having equivalent impact to the more traditional technologies of lithography, STM-based manipulation, and self-assembly.

C. Temporal Control

Surface chemistry can be induced by tunneling electrons from STM, photogenerated electrons from a fs laser, and particles (ions, atoms, molecules, clusters). Several different mechanisms are known to be important in these interactions. For example, a

mechanism which involves inelastic negative ion resonance scattering is often responsible for the induced reactions. Using fs lasers, the goal is to probe the time scales for bond breaking and formation on the surface. Since the photons are confined to a pulse of 10 - 100 fs duration, the induced chemistry can be significantly different than that induced by cw and ns laser and particle pulses. Excitation with fs laser pulses also allows the study of energy transfer and coupling of the electronic degree of freedom to the nuclear motion. It is very useful to consider fs induced chemistry of the same molecules in the gas phase and in solutions, which provides a general understanding of chemical dynamics. In excitation with fs laser pulses, important information comes from using an excitation time scale that is shorter than, or comparable to, the time scales involved in energy relaxation and transfer. By stretching the laser pulse to a ps timescale, the excitation rate becomes smaller than the relaxation rate, resulting in different reaction pathways. The combination of fs lasers with STMs provides an opportunity to probe, manipulate, and control matter on the fs temporal and angstrom spatial scales. Nonlinear optical excitation is expected at a tunnel junction, resulting in the measurement of atomically resolved optical properties.

IV. Other Advanced Techniques for Probing Particle- and Photon-Surface Interactions

In addition to those mentioned in earlier sections of this report, several unique new techniques have been developed over the past decade, many of which are still evolving, that offer unique opportunities for probing particle-and photon-surface interactions. Many of these new techniques have strong overlap with existing AMOP interests and expertise.

A. Beams of Ions, Atoms, Molecules, Clusters, Electrons, and Photons

The use of highly focused beams to study surfaces is of interest for several reasons. A focused beam can deposit more energy or material in a small spot, thus stimulating changes in the local effective temperature or concentration of reacting/exciting species. Such beams can also be rastered over the surface (either by moving the beam or by moving the sample) so as to yield laterally-resolved images of the surface. Such imaging is becoming increasingly more important for complex, technologically or environmentally relevant surfaces that are often highly heterogeneous in the lateral dimensions. Rastering can also be used to write nanometer-scale patterns on surfaces in a variety of materials, thus presenting exciting possibilities for the characterization and production of next-generation semiconductor circuits and magnetic storage devices. By combining imaging while sputtering, it is possible to carry out 3D tomography of solids of arbitrary composition at an unprecedented resolution. Commercially available ion beam sources permit focusing of Ga^+ ions at 15-50 keV down to a spot size approaching 5 nm. With further development using AMOP techniques, it should be possible to improve on this resolution and/or create beams of state and energy selected ions at lower energies. The sizes of such beams are thus smaller than the collision cascade induced in the solid by the projectile itself, opening up the possibility of studying the physics of the cascade process in more detail than has been possible previously. With associated theoretical interpretation, such studies would have a very positive impact on the technique of secondary ion mass spectrometry that is an ubiquitous tool in materials science and, particularly, in the electronics industry. These focused probes, when combined with mass spectrometry, offer new possibilities for high-spatial-resolution chemical imaging with applications in geology, environmental science, and biology.

Photon beams from third-generation light sources can be focused down to 10-20 nm, permitting simultaneous spectroscopy and microscopy (via sample scanning). Such “spectromicroscopy” can be fruitfully applied to atomic and molecular species deposited on surfaces, e.g. from ion or atom beams. Using both focused ion beams and focused photon

beams together would permit studies of the rates of diffusion of species as well as changes in chemical state with lateral displacement away from the position of the primary collision. Core-and valence-level photoelectron spectroscopy, Auger electron spectroscopy, and soft x-ray absorption spectroscopy can be carried out with unprecedented resolution and experimental control, including the variation of light polarization and measurement of outgoing electron spin.

Optical pumping or magnetic state selection via hexapole magnets can produce beams of aligned atoms and ions. Studies of spin dependences and their surface interactions will be invaluable for probing of magnetic surfaces and interfaces, giant magnetoresistive structures, colossal magnetoresistive compounds, and high-temperature superconductors.

B. Advanced Combinations of Supersonic Beams, Atomic Traps, and Atom Optics

Supersonic beams have revolutionized our understanding of precision spectroscopy and reaction dynamics. Atomic traps have had a similar impact in many fields, spanning the range from atomic fountains for clocks to Bose-Einstein condensation. Atom optics are now being applied for precision measurements, interferometry, and pattern generation including holography. It is clear that outstanding scientific and technological advances will result from the combined use of these incisive techniques. New techniques involving high-brightness neutral beams, spatially-guided and tightly-focused neutral beams, and mesoscopic pattern deposition will follow from such studies. Already extant exploratory studies involving ultra-cold and spatially controlled neutral beams injected into optical fibers may lead to new classes of atomic gyroscopes. Atoms exiting such fiber-optics based devices may find application as atomic "fountain pens" for lithography and mesoscale chemistry.

C. Femtosecond (fs) Surface Spectroscopy

The development of ultrashort laser pulses that are now typically 60 fs in duration, but with values as short as 5-10 fs having been obtained, has opened up new possibilities for studying electron, photon, and energy transfer dynamics at surfaces. In these experiments, single- and multi-photon absorption can lead to time-resolved electron excitation, e.g. into image states near surfaces that are one-dimensional analogues of Rydberg states in atoms. Electron emission from these excited states can be studied by using pump-probe techniques. Thus, the energies and lifetimes of electronic states near surfaces can be probed as well as the influence of photochemically-induced surface reactions. Alternatively, the energy from a single laser pulse can be deposited over a short time into secondary electron creation, with these electrons then driving surface chemical bond changes that can be followed in time, e.g., via time-of-flight mass spectrometry.

D. Spatial- and Temporal-Imaging of Surface Atomic Structures

New probes of surfaces based on ion and photon beams also offer the possibility of studying the internal atomic structures of surfaces on time scales relevant to surface chemical processes. For example, in the recently developed technique of scattering and recoiling imaging spectroscopy (SARIS), the atomic structure is determined via two-dimensional ion scattering patterns from a crystal in a time-of-flight system. The pattern acquisition time is in the few-second range and individual images of the scattered and recoiled particles can be resolved on a 10 ns scale. For many surface reactions at near-UHV conditions, this is rapid with respect to surface atomic diffusion and reaction. These ion scattering images exhibit extreme sensitivity to surface structure, providing a real-space, element-specific surface crystallography. This technique also offers new opportunities in the detection of surface hydrogen, the study of non-planar scattering events, the analysis of disorder at surfaces, determination of surface diffusion and kinetics, and the

study of ion-surface charge transfer characteristics. New applications of this ion scattering technique to liquid surfaces, which are currently emerging, offer the possibility of monitoring dynamics on fluid surfaces. With high-brightness photon beams from third-generation synchrotron radiation, it is currently possible to measure high-resolution chemical-state-resolved core-level photoelectron spectra on time scales in the second range and to measure two-dimensional photoelectron scattering and diffraction patterns from the crystal lattice in minutes. Similar information can be obtained on a smaller scale by Auger mapping with finely focused electron beams. Such ion and photon probes can follow the development of different chemical states in time, thus directly deriving kinetic information. The scattering images and diffraction patterns also permit determination of local atomic structures for the various chemical states present. Such measurements could be applied, for example, to surface reactions carried out with state-selected ion or atom beams.

E. Coincidence Techniques

Coincidence measurement techniques are widely used in AMO physics. Application of similar techniques to surface studies offers several advantages. The use of pulsed ion, atom, or photon probe beams coupled with coincidence measurements of outgoing ions or electrons, or among the various products of the excitation, could significantly enhance our knowledge of energy flow and time evolution in surface processes, including the electronic states and atomic configurations involved in desorption, decomposition, and electron emission. One challenge in such measurements is that surfaces are copious sources of secondary electrons, ions, and atoms, making the detection of true coincidences more difficult. Other problems are the difficulty in efficiently collecting ions or electrons from surfaces and the ability of surfaces to reneutralize and recapture species. Electron-electron and electron-ion coincidence measurements are currently being studied by a few research groups. Expansion of such efforts would provide new insights into the details of dynamic surface processes.

F. Surface Studies in High Pressure Environments

Most surface studies have been carried out in ultrahigh vacuum environments, with possible pretreatment of the surface at slightly higher pressures so as to carry out chemical reactions. The ability to carry out measurements at higher effective pressures of up to even 10^{-3} torr would permit duplicating the conditions found in semiconductor device and magnetic disc fabrication and provide data of more technological relevance. Optical probes are most readily adapted to high pressure environments since photons can travel through dense gases under appropriate conditions. An example is probing second-harmonic generation with lasers, fluorescence, and Raman techniques. Particle beams present a greater challenge since they are attenuated in dense gases. The use of particle beams could be achieved by using ionic or atomic beams of suitable flux and/or surrounding the sample with a differentially-pumped cell whose apertures accommodate the entrance of the excitation source and the exit of the detected products. With suitable designs, it might be possible to work at pressures in the 5-10 torr range, thus more nearly approaching the conditions of atmospheric and environmental reactions on surfaces.

G. Higher-Efficiency and Higher-Speed Detectors

Developments in particle and photon detectors now allow single particle detection under a wide range of conditions. However, this is not yet possible in some important cases, such as very large molecules and clusters ($>10^6$ Daltons) in mass spectrometry, atoms and molecules sputtered from surfaces, and low energy atoms and molecules important in new processing applications. The development of higher-speed and higher-efficiency detectors for atoms and ions requires an understanding of how the kinetic energy of atomic motion is transferred to electronic excitations in solids

at low impact velocities, resulting in electron emission. Even with more intense beams of both photons and ions, many experiments are detection-rate limited. For example, the rate of acquisition of photoelectron spectra is currently limited by the capabilities of multichannel detectors. For ions or atoms, detection is complicated further by the low and strongly energy dependent efficiency of the detectors. The standard particle detectors employed in particle/surface interaction experiments are the channeltron multiplier and the microchannel plate array. These have near unity efficiency for detection of electrons and energetic ions at 10 keV, but the efficiency for ions or atoms monotonically falls to zero as the velocity of the ions decreases. Developments in detector technology would enormously expand the capabilities of many beam scattering techniques, permitting more sensitive and/or higher-resolution studies in many surface characterization and surface analysis studies. Possible approaches for enhancing particle detection involve laser ionization of the neutral beam and/or development of new surface coatings with higher electron emission coefficients. Fundamental research on detector development, including how the kinetic energy of atomic/molecular motion is most efficiently transferred to electronic excitations in solid surfaces at low impact velocities, would thus be very beneficial.

V. Key Research Areas in This Report That are Under-Represented in the United States

- Basic studies of the interactions of low energy (0.1eV - 0.5 keV), well-defined neutral and ion beams with surfaces is vastly under-represented in this country. Germany, Japan, France, and The Netherlands are presently leading the ion beam field. The area is replete with important applications.
- Basic studies of the effects of low energy (1 eV - 1 keV) electron beams with surfaces are under-represented in this country. This area also has important applications.
- The STM was developed in Europe and novel applications of scanning probes are continually being demonstrated, especially in Germany and Switzerland.
- Both Japan and Germany have substantially larger commitments to femtosecond laser work in chemistry, biology, and materials science.

VI. Summary List of Basic Unsolved Problems Involving the Interactions of Low Energy Particles (Electrons, Ions, Atoms, Molecules, Clusters) and Photons with Surfaces

1. Theoretical treatment of excited states at interfaces.
2. Atomically resolved structure determination of surfaces and adsorbed species.
3. Quantum-state-specific control and atomically- and chemically-resolved spectroscopic characterization and manipulation of surfaces.
4. Time- and spatial-resolved spectroscopic monitoring of dynamic bond breaking and formation processes at surfaces.
5. Dynamics of energy and charge transfer at surfaces, i.e. the flow of energy and charge and the motions of atoms in particle-surface interactions.

VII. Summary List of Applications of Particle and Photon Interactions at Surfaces

1. Nonequilibrium growth, nanoscale fabrication, and processing of materials.
2. New techniques involving high intensity, spatially guided neutral and ion beams for mesoscopic pattern deposition.
3. Mechanisms of surface chemical reactions and catalysis.
4. Environmental problems, including sensing and processing of biological molecules.
5. Futuristic materials based on atomic and molecular manipulations.

VIII. Conclusion Statement

This report has identified some important research needs, opportunities, and applications that can be achieved through the interaction of two groups of scientists, i.e. atomic, molecular, and optical physicists and surface scientists. The synergistic expertise and techniques from both groups provide an unparalleled opportunity to make significant advances in our basic understanding and application of the interactions of energetic particles (electrons, ions, atoms, molecules, and clusters) and photons with surfaces.

Panel D: Theory of Structure and Dynamics

Chair:
Chris Greene (University of Colorado)

Panelists:
Alexander Delgarno (Harvard Smithsonian Center for Astrophysics)
Charlotte Fischer (Vanderbilt University)
Kate Kirby (Harvard College Observatory)
Kenneth Kulander (Lawrence Livermore National Laboratory)
Uzi Landman (Georgia Institute of Technology)
James McGuire (Institut fur Kernphysik)
Vincent McKoy (California Institute of Technology)
Anthony Starace (University of Nebraska)
Malcolm Stocks (Oak Ridge National Laboratory)
David Yarkony (Johns Hopkins University)

Executive Summary

In AMO physics, the terms "structure and dynamics" encompass diverse topical areas that intellectually challenge and relate closely to the mission of the Department of Energy (DOE). In these working papers, we summarize our findings. Our deliberations have identified key themes in the theory community that overlap and further the research goals of Basic Energy Sciences at DOE, broadly-construed. This panel has assessed new areas ripe for future development within the next 5-10 years. Because our charge has been specifically aimed at identifying new research directions, we stress at the outset that many exciting theoretical (and experimental) areas will not be explicitly discussed. Other theoretical areas with great potential and relevance to the DOE mission are not covered in this report. We have no room, for example, to detail the varied and deep connections of AMO science to the disciplines of astrophysics, atmospheric science, space science, environmental science, nuclear physics, particle and fundamental physics and the physics of reactive flows and shock waves.

The areas of greatest excitement and potential for near-term development in structure and dynamics are grouped into five categories: Control; AMO physics in extreme conditions; Novel states of matter and light; Extending the frontiers; Opportunities and horizons in high-performance computing. We begin with a short summary of each of these categories. The rest of our report elaborates on each in somewhat greater detail. A continuing theme in all of these areas is a natural progression of knowledge and understanding from simple prototype atomic species to more complex species like molecules, clusters, and surfaces.

"Control" is an important theme for the future. It embodies a clear long-range goal for AMO science. The field of coherent control, for instance, involves the manipulation of atomic and molecular processes by the design of femtosecond light pulses that cause the system to respond in a prespecified manner. Just as the development of femtosecond lasers has brought coherent control to the brink of reality, so has the enhanced ability to manipulate atoms on surfaces brought the potential of engineered nanostructures to the forefront of this field. The techniques of atom interferometry and lithography will certainly play key roles in the design of nanostructures.

"AMO physics in extreme conditions" includes unusual environments where basic scientific questions remain inadequately understood. Atomic and molecular collisions at ultracold temperatures, for instance, probe the pure quantum limits. Ultra-intense laser fields produce dramatic new physical effects and processes, while ultra-short light pulses are important for the *"Control"* goals discussed above. New generation synchrotron light sources probe atomic and molecular structure at ultra-high resolution (and in some cases at ultra-high energies), which magnifies the limitations of conventional independent-particle models and points to the need for improved theoretical capabilities.

"Novel states of matter and light" have energized much of the scientific community in a fast-paced exploration of striking new directions, largely driven by recent experimental progress. Several groups can now create BEC in their laboratories on a daily basis, but at the same time many basic theoretical issues require a far deeper understanding. Quantum optics theory and experiment, especially in the area of laser cooling of atoms, continue to show rapid, exciting progress.

"Extending the frontiers" groups together several areas that touch on or encompass current work in the BES AMO Physics program. New directions and current bottlenecks deserving of priority effort are mentioned, including the direct description of real-time dynamics for problems like intense laser-atom interactions, where the interaction is strong and localized in time, and must be treated nonperturbatively. Another area ripe for extension is the theoretical study of relativistic correlation effects,

both in heavy atoms and in simple molecules containing heavy atoms. These tie in with the DOE mission, as discussed below. Short-time dynamical studies using periodic orbit theory and scaled-variable spectroscopy can be applied to a range of new problems. Finally, the interpretation of energetic plasmas requires a deeper understanding and the development of methods to explore a large number of processes involving collisions of electrons, ions, atoms, and molecules.

"Opportunities and horizons in high-performance computing" includes areas where progress in AMO science hinges on the utilization of large-memory, scalable multicomputers. The high performance of these powerful modern computers can dramatically enhance our ability to achieve robust descriptions of fundamental processes and complex systems across a wide spectrum of AMO science. Development of scalable and paradigm-changing algorithms required for efficient use of these new computing architectures, coupled with ready access to large parallel computers, can lead to major breakthroughs in computationally intensive applications and can enhance the impact of theory on experiment.

Elaboration on the Five Themes Described Briefly Above

Before we discuss the scientific directions and connections identified by our panel, we believe it is important to recognize one of the constraints we faced in our undertaking. The separation of "Theory" into one panel at this workshop, split off from the other topical areas represented in this report, is unfortunate because the most productive advances in science—especially in AMO science—result from fruitful synergism between experiment and theory. Consequently, most of the forefront areas mentioned in this section overlap closely with the areas discussed by the other panels. In cases where the other panels propose an area ripe for further development in the coming decade, it should be understood that extensive theoretical contributions can be very valuable for guiding and interpreting experiments. However, theory does differ from experiment in "equipment needs." Access to large-scale computational hardware is needed by theorists in order to undertake certain types of cutting-edge calculations.

CONTROL

Coherent control

The dynamics of quantum systems can be modified by coherent light pulses, and energy can be moved around in a predetermined way on an atomic scale. Advances in short pulse laser technology have made it likely that the long-sought goal of quantum control of molecular collision dynamics will finally be achieved. In principle, manipulation of a time-dependent pulse will permit the subsequent evolution of the system to be controlled. Amplified few-fs Ti:sapphire pulses have focused intensities larger than 10^{14} W/cm^2, which is strong enough to drive molecular transitions efficiently. Selective dissociation, control of reactions, and preparation of desired molecular states or structures can be achieved by these methods. Accomplishing these goals requires intimate interaction between experiment and theory. Knowledge of the molecular potential energy surfaces, dipole coupling strengths as functions of molecular geometry and the ability to determine the variations of the time evolution of the coherent wave packet caused by changes in the phases of the pulse will allow optimization of the product yields. Precise determination of these required parameters exceeds existing theoretical capabilities for all but the simplest diatomic systems.

For more complicated systems, nevertheless, theory can provide approximate starting points for experiments designed to provide feedback to more accurately calibrate the system response, leading iteratively to the desired results. One anticipated experimental problem is that the UV or VUV wavelengths required to induce an electronic transition in most molecular systems are much shorter than those produced by the Ti:sapphire amplifier. Harmonic conversion or frequency mixing techniques can

generate coherent light pulses, as short as (or even shorter than) the incident pulse, with almost any central frequency. Coherent control provides the capability to probe and understand, at the most fundamental level, the making and breaking of chemical bonds in addition to enabling the production of novel product states of the quantum system. This can mean the generation of new chemical compounds, the establishment of more efficient routes to desired reaction products or even the construction of particular structures of the product molecules. This technology has applications beyond the modification of molecular collision dynamics, such as the generation of Rydberg wave packets in atoms and the control of current flow in semiconductors.

Nanoscale Materials: Clusters, Nanocrystals, and Dots

Investigations of the microscopic physical origins underlying size-evolutionary patterns of materials properties are among the main themes in modern condensed matter physics and materials science. These studies are enabled by the emergence of novel experimental probes and by the development of new theoretical and algorithmic approaches. Coupled with the availability of high-powered computational tools, they allow deep insights into the structure of matter bridging the atomic, molecular, and condensed-matter regimes. In addition to their basic scientific significance, the above issues form the scientific base for technological developments, particularly in the area of device miniaturization.

Atomic material aggregates with nanometer-scale dimensions exhibit certain properties analogous to those found in other "zero-dimensional" (0D) systems, namely atoms and nuclei. Most prominent among these is the electronic shell structure of small (metallic) clusters. At shell closures this leads to enhanced stability and self-selection of clusters having "magic numbers" of atoms. Moreover, other properties such as ionization potentials, electron affinities, and single-atom detachment energies of open-shell clusters exhibit odd-even alternations (as functions of the number of atoms). These originate from cluster shape deformations similar to the Jahn-Teller distortions familiar in molecular (and nuclear) physics. Such clusters possess collective excitation spectra (giant photoabsorption resonances), and they fission upon multiple ionization.

The adaptation of AMO physics methodologies and their application to clusters and nanocrystals, in conjunction with computer-based (time-dependent) first-principles molecular dynamics simulations, could contribute significantly to elucidation of the above-mentioned phenomena. Work in this area also bears on problems pertaining to: supershells and semiclassical descriptions (using periodic-orbit theory) of electronic spectra for larger clusters; multiple charging (shape resonances), basis-set selection and correlation effects; single-particle and collective (plasma) excitations; atom (or ion)-cluster and intercluster collision dynamics, product branching ratios, and collisional energy redistribution; cluster and nanocrystal growth mechanisms; cluster thermodynamics (structural and phase transformations); magnetic properties; dipole (and higher multipole) excess electron binding to molecular clusters [e.g. $(H_2O)_n^-$, n>2] and solvation energetics and dynamics; and size-dependent reactivity. These studies could lead to formulation of atomic-scale principles to guide the controlled design and preparation of nano-scale materials with prescribed size, shape, property, and functionality.

Another class of novel 0D nanoscale structures, with close analogies to atomic systems, consists of confined (2D) electron gases in the form of dots. These are often referred to as "super-atoms". Both basic science and technology will be advanced by development of an understanding of the electronic level structure, of the excitation spectra (both single-particle and collective resonances), and of the charging properties in such systems. The improvement of theories beyond the mean-field approximation is essential, both under field-free conditions and under the influence of an applied magnetic or optical field.

Atom Interferometry and Lithography

Atom optics has shown promise in recent years, both for the fundamental interest it has generated and for its technological promise. Atom interferometry has progressed beyond the demonstration of basic two- and multi-slit interference patterns used as textbook illustrations of quantum mechanics. It can now be used as a tool to measure atomic properties including the *phases* of quantum amplitudes in addition to their magnitudes. The future applications of these interferometric techniques are hard to envision fully at this time, but they will certainly probe the quantum mechanical ideas of entanglement, coherence, and decoherence. Experimental work in this area would benefit from theoretical studies, particularly when noise and dissipation play an important role in the interference structures.

As technologies continue to improve for handling phase-coherent atomic beams of small dimension, so will the ability to lay down atoms on surfaces in prescribed arrangements. The continued development of these novel lithographic techniques is likely to have far-reaching application to microchip production and nanoscale fabrication.

AMO PHYSICS IN EXTREME CONDITIONS

The theoretical description of AMO phenomena under extreme conditions generally requires the development of appropriate (and often new) theoretical approaches. Advances in experimental technologies have permitted measurements covering a broad range of such phenomena, thus providing the opportunity for theory and experiment to progress together. Many of these opportunities have significant applications to energy-related technologies. The following are some of the leading opportunities:

Ultra Cold Collisions

At sub-milliKelvin temperatures, atom-atom collisions enter a new regime in which de Broglie wavelengths are far larger than atomic dimensions and in which collision times are longer than atomic radiative decay times. Atomic collision cross sections are dependent on long-range atom-atom interaction potentials. At the lowest energies, hyperfine interaction effects must be included. Acquisition of a deeper understanding of these cold collisions is crucial to the control and application of BEC phenomena, to improvements in the precision and stability of atomic clocks, and to enhancements in the sensitivity of experiments to detect small effects and to make precision measurements.

Ultra Intense Fields

At laser intensities greater than about 10^{12} W/cm^2, nonlinear laser-atom effects (e.g., above threshold ionization, high harmonic generation, etc.) become increasingly prominent; above about 10^{19} W/cm^2, the electric field in the laser light—rather than the nuclear Coulomb field—controls electronic motion and the induced electron velocity becomes relativistic; above about 10^{29} W/cm^2, the laser field is strong enough to "spark the vacuum," i.e., to create electron-positron pairs. The theoretical description of such phenomena requires a non-perturbative treatment of the laser-atom interaction and, often, a time-dependent solution of the quantum equations for the laser-induced electronic motion. Prominent among possible applications is the use of high harmonic generation to produce coherent sources of VUV and X-ray radiation.

Ultra Short Pulses

The development of laser pulses as short as 5 fs is now possible, and shorter pulse durations are on the immediate horizon. Pulses this short permit excitations of localized electronic or vibronic wavepackets in atoms and molecules. The theoretical description of the electronic dynamics requires a time-dependent approach. Furthermore, additional coherent, short laser pulses permit one to control the evolution of the system. Analysis of the wavepacket motion in atoms or molecules has important applications. In Rydberg atoms, theory can explore the connections between

classical and quantum behaviors. In molecules, a goal for theory and experiment is to develop robust methods for controlling the evolution of the probability density along particular desired Born-Oppenheimer pathways.

Ultra High Resolution and Detection Sensitivity

Lasers and third generation synchrotron light sources are permitting atomic and molecular spectra to be observed with unprecedented resolution and sensitivity. Energy resolutions of 1 meV and better are becoming available in the 100 eV photon energy range; while this does not translate into a "resolution" figure as impressive as can be achieved with state-of-the-art CW laser systems, it is a major step forward in the high energy regime. Cross sections as small as hundredths of a barn are being observed. In molecules, vibrational and even rotational energy levels can now be resolved even in some synchrotron experiments. In atoms and in atomic negative ions, such high energy resolution and sensitivity requires theory to transcend the independent-electron model to incorporate many-body correlation effects, as well as the effects of normally feeble interactions (e.g., polarization effects, spin-orbit and other relativistic effects). In molecules, theory at an elementary level has not been widely applied, even for relatively small systems. Photodetachment of negative ions leads to spectra dominated by correlated two-electron resonances, with spectral patterns very different from the one-electron Rydberg series omnipresent in neutrals. Knowledge about such resonance features is essential for a number of energy technologies, such as in the design of gas lasers, arcs, fluorescent lamps, and high-power discharge switches, because resonances affect the conductivity of a low-temperature discharge and plasma environment.

New experimental techniques hold promise also in collision physics as they allow more detailed analysis of chemical kinematics and reaction dynamics. One such technique is cold target recoil ion momentum spectroscopy (COLTRIMS). Analyses of data obtained with this method have used classical methods, but analyses using detailed quantum calculations are largely incomplete. This is true not only for many-electron systems but also for simple systems at low velocities. A difficult challenge for quantum theory is to develop methods that go beyond the independent particle model for reactions involving multiple electron transitions. Often these many-electron transitions are dominated by the dynamics of electron correlation, which is a key to understanding mechanisms for energy transfer in complex atomic systems. The general question is how to understand complex systems (including large molecules or nanostructures) in terms of simpler atomic systems.

Ultra Fast Collisions

An example of a highly localized conversion of energy occurs in interactions of relativistic ions with atoms in which electron-positron pairs are produced. Calculations have not yet been able to give a reliable interpretation of the spectra observed, in which it is believed that disruption of the negative energy sea has occurred.

Ultra High Photon Energies

Another window of opportunity is being opened by new third and fourth generation synchrotron radiation facilities. These facilities are just beginning to produce data useful in both the gaseous and condensed phases of matter. For example, Raman emission by x-ray scattering is being used to determine composition of materials used in new semi-conducting devices. Interpretation of these experiments requires a theorist to consider both atomic and condensed-matter issues. The development of a microscopic understanding of the effects of high energy photons that penetrate matter requires us to unravel multiple ionization and excitation events produced by x-rays, both in photoionization and in Compton scattering. Cross sections and reaction rates for these processes are dominated by the concerted motions of non-independent electrons, and photons probe the electron correlation dynamics cleanly. Sorting out such processes could help us to understand how to modify

or control site-specific reactions in more complex chemical and biological systems.

NOVEL STATES OF MATTER AND LIGHT

Bose-Einstein Condensation (BEC) and atom lasers

Interest in AMO science intensified in 1995 following the first experimental observation of BEC for a dilute gas of spin-polarized alkali-metal atoms. The ensuing excitement has had few parallels in the physical sciences. Experimental laboratories worldwide quickly initiated their own attempts to reproduce the first experiments, and at last count six independent groups had achieved BEC. While the initial observation was primarily an experimental achievement, theoretical studies have since played a key role in the continued growth and excitement of the field. BEC theory is now a high profile activity represented in some of the best physics departments, and is carried out by both condensed matter and AMO theorists. This subject has already spawned extensive interdisciplinary interactions that extend our understanding of the many-body problem from the perspectives of both fields. Fundamental questions arise relating to: the mechanisms of spontaneous symmetry breaking and the formation of coherence; phenomena such as superfluid flow and persistent currents; specific heat and critical temperatures; first and second sound waves and excitations; the kinetic and dynamic evolution of the condensate.

This panel believes that theoretical efforts to understand BEC will help to unravel a number of issues that need to be better understood such as: the energy spectrum of this many-body condensate; its formation and decay mechanisms; its coherence properties; the variety of novel forms in which it can be created. A key question concerning its possible relevance to DOE interests arises from the simple energetics of BEC. What is usually termed the "ground state" of the condensate is not actually the true ground state of this many-body system; it is a metastable state whose total energy is several electron volts per atom higher than the ground state. In other words, the condensate, despite being ultracold (50nK) translationally, stores a surprisingly high density of internal energy since about 10^6 atoms are confined to a volume of order 10^{-18} m^3. Another type of proposed condensate consists of helium atoms in metastable electronic states; if experiments succeed in forming such a condensate, the amount of energy stored per atom would be increased by nearly an additional order of magnitude. Should future research demonstrate ways to achieve the controlled release of this energy, e.g. through an "output transducer," this could be an important source of coherent atoms. Such a phase-coherent beam of atoms was demonstrated earlier this year at MIT, and has been called an "atom laser." The atom laser beam is qualitatively different from an ordinary incoherent atomic beam, and is likely to spawn spectacular capabilities for lithography and atom interferometry in the coming decade.

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Quantum Optics

Entanglement: Quantum optics brings fundamental quantum mechanical theory to bear on thought experiments that have recently been realized experimentally with the advent of high-precision cavities and state-specific control of atomic states through stable, tunable lasers. Micromaser cavities with exceptionally high *Q*-factors provide photon traps, analogous to particle traps, which are used to localize injected photon fields. These fields are typically tunable over a wide range of frequencies near resonance between two atomic levels of interest, e.g., alkali Rydberg levels. The passage of one or more atoms through such a cavity leads to an entanglement of the atom with the cavity, which can then be probed by a second atom, which in turn becomes entangled with the first atom. Alternatively, two cavities can be entangled with one another via the passage of a single atom, which might transfer a photon from one cavity to the other. All of these coherent processes are manifestly quantum mechanical in nature, as is revealed dramatically when the states are correlated

with macroscopic detectors. Note that the use of explicitly nonclassical light fields (i.e. squeezed states) for high-precision frequency metrology is presently funded in the DOE Atomic Physics Program. Related fertile areas include:

Dissipation theory: A central theme in quantum optics has been the development of powerful theoretical methods that can efficiently handle irreversibility in nonrelativistic quantum mechanics. These include quantum Monte Carlo and quantum trajectory methods in which the continuous evolution of the Schroedinger equation is disrupted by quantum jumps of the state. The concepts are general and diversely applicable in quantum mechanics. In many-state simulations they are far more efficient than the more standard "master equation" techniques.

Quantum computing: When the usual binary logic of computation is modified by quantum mechanically entangling two states, this leads to exponential gains in computation speed for certain important problems. The experimental difficulties arising from decoherence for realizing a quantum computer are formidable, but the potential gains are extremely high. Proposals for quantum error correction, transfer and cloning of quantum states, quantum communication, and quantum cryptography are all interesting by-products of this research. It has been shown that arbitrary quantum states can be engineered, and using recently-developed tomographic methods, experiments can characterize these states completely.

Laser cooling and atom optics: The interaction of atoms with the electromagnetic field allows trapping and cooling and to create optical elements such as beam splitters and mirrors for matter-wave interferometers. There have been tremendous experimental advances in laser cooling techniques over recent years with the lowest temperatures given by dark state cooling in which the coldest atoms become decoupled from the field through a destructive internal interference of the absorption amplitude. Atoms cooled in this way undergo Levy flights and have interesting and anomalous statistical properties. Experiments on lattices of light that confine atoms in a three-dimensional array of optical potential wells are important for precision measurements.

Antimatter and exotic species

Experimentalists and theorists have recently studied the spectroscopy of antiprotonic helium. Efforts continue toward creating antihydrogen at low energies for tests of fundamental physics. A dedicated antiproton storage ring is being built at CERN, which will enable the collection of antiprotons in a Penning trap. As these experimental capabilities leap forward, the branch of atomic physics concerned with the structure and collisions of exotic particles at low energies has many opportunities and challenges, in which theoretical contributions are essential. Enormous amounts of energy could be stored in the form of antimatter, if its creation and manipulation advances tremendously beyond present-day capabilities, though this appears too remote to serve as a goal for the immediate future.

EXTENDING THE FRONTIERS

Time-Dependent Real-Time Dynamics

The dynamics of quantum systems can often be described directly in the time domain. This approach provides accurate product-state distributions over a range of total energies from a single calculation. When coupled with modern visualization tools, it can provide key insights into the underlying mechanisms. For many processes this is the only practical formulation, especially in cases for which the number of open channels is very high (e.g. when three-body continua are present) or where the Hamiltonian for the system is explicitly time-dependent (e.g. when an intense laser pulse is applied). For even the simplest systems this approach can require very substantial computational resources. Utilization of large multi-processor computers has allowed the investigation of many phenomena for simple systems that are central to the missions of the DOE. Many of the currently-used methods solve the time-dependent Schroedinger equation iteratively—a scheme that is

ideally suited to parallel architectures. The complexity of problems that can be addressed in the future is contingent upon the development of efficient algorithms that maximize the division of labor among numerous fast processors. Problems that have been successfully investigated include molecular dissociation by electron, ion, or photon impact; inelastic and reactive atomic and molecular scattering processes; atomic excitation and ionization by intense, pulsed laser fields.

An outstanding problem in this area is the study of multielectron systems subject to a time-dependent interaction: either a collision or a pulsed electromagnetic field. Even for two electron systems, there are many problems that have not yet been solved. The independent-particle approximation often fails dramatically in a time-dependent problem, and is thus inadequate. Resources and numerical methods are presently being developed that should eventually permit an accurate numerical solution of the full-dimensional two-electron problem for a number of non-trivial regimes. Among these is the surprisingly strong simultaneous double ionization of helium by a short pulse optical laser field: interpretation of these experiments continues to resist theoretical efforts. The calculations challenge our ability to solve the time-dependent Schroedinger equation, of course, but perhaps of equal importance, they challenge our visualization skills needed to identify the dynamical mechanisms that operate. A desired outcome is that, based on the insight gleaned from a solution of the dynamics for two-electrons in a laser field, a general improved technique will be developed that can handle similar nonperturbative problems for multielectron systems.

Relativity and correlations in structure and dynamics

Two active research areas that have been traditionally addressed within current AMO theory projects in this program include *electron correlations* and *relativistic effects*. The study of correlations aims broadly at the derivation of theoretical tools capable of describing numerous atomic phenomena that are beyond the scope of the independent-particle approximation: the binding of an electron to heavy atoms with closed subshells; the escape of two or more electrons following absorption of a single photon; collective excitations of two-electron resonance states. Numerous successes of this line of research can be identified. Theoretical capabilities have advanced tremendously, especially in the last few years. While this field maintains a continuing interest in further improving theoretical methods for even the simplest atoms with only two or three valence electrons, we focus here on new advances that are increasingly desirable. This is truly a frontier, a theoretical subfield that remains, comparatively speaking, in its infancy: the description of electron correlation effects in bound and continous spectra where relativistic effects are important. This includes mainly the atoms heavier than, say, Kr (Z=36), as well as diatomic and triatomic molecules containing one or more of these heavy atoms.

In the atomic realm, the most commonly-used approaches start with the Dirac-Coulomb Hamiltonian, and a multiconfiguration Dirac-Fock wave function expansion which, in the case of fixed basis set methods can be obtained through configuration interaction (CI) calculations. Further effects can be added perturbatively. Despite the ability to treat aspects of the low-lying bound state spectrum for far more complex open-shell atoms and negative ions than was conceivable ten years ago, roadblocks loom. To be sure, the spectra of these species are extremely complicated, especially in the lanthanides (currently important for the lighting industry) and in the actinide and transuranic elements (of continuing relevance to the DOE environmental cleanup mission). For example, in Lr (Z=103), theoretical investigations have concentrated on the determination of the ground state properties, but even the ionization energy is not known accurately. The number of configurations that arise, even in the most constrained CI expansions, grows explosively in these systems with one or more open subshells. In fact, it is worth asking whether calculations based on this methodology truly have a realistic hope of being able to calculate accurate

energy levels, photoabsorption and photoionization oscillator strengths, dielectronic recombination, and hyperfine and weak-interaction couplings, at a level of precision that is spectroscopically useful. Multichannel level perturbations are so rampant that it may make sense to pursue the more limited initial goal of describing short-time spectra, i.e. the spectroscopy at a comparatively coarse-grained energy resolution.

Many of these same bottlenecks arise in the theoretical description of a diatomic or triatomic molecule, of course. Additional complexities arise because of the much greater difficulty of performing an electronic calculation in a multicenter geometry, and with the intimidating complexity of the nuclear dynamics on nonadiabatically-coupled potential surfaces. In triatomics, conical intersections lead to all the difficulties, e.g., of Jahn-Teller effects that modify the molecular symmetry one would expect based on fixed-nuclei calculations. Atom-diatom scattering calculations can sometimes describe these effects with modest computational effort, and in a conceptually-appealing manner, by using geometric phases appropriately. Moreover, while the construction of bound-electronic potential surfaces and a description of the nuclear dynamics is a tremendous challenge, one would like the theory to advance and develop the capability to treat electronic continua that arise in photoionization, photodetachment, and electron scattering processes. In the molecular realm, much remains to be understood even for molecules containing the first-row transition metal atoms, where relativity can still be handled perturbatively.

The research in this area requires extensive conceptual and computational development. We believe that it is poised to make tremendous strides in the coming decade, by coupling the deeper understanding we have achieved in how to treat these types of bound and continuum problems with the rapid improvements in computational hardware and algorithms.

Quantum Mechanical Approximations Derived from Semiclassical Methods

A minor revolution has occurred during the past decade in the theoretical description of quantum systems with two or more strongly-coupled degrees of freedom. The most exciting possibilities arise in systems whose classical Newtonian analogues are chaotic. Classical chaos arises when classical trajectories are exponentially sensitive to initial conditions. The quantum dynamics for such systems is sometimes referred to as “quantum chaos.”

Our panel has for the most part avoided a discussion of theoretical possibilities based on the merits of any particular “technique.” The semiclassical “quantum chaos” methods devised by Gutzwiller, Berry, and their followers seem to us worth singling out, however, because their new capabilities complement more conventional schemes used by AMO theorists. In particular, new types of spectroscopies (such as scaled-variable spectroscopy) give deep insight into the origin of short-time features in the “time domain” of the observables such as photoabsorption cross sections. Of course, short-time features translate via Fourier analysis into global, coarse-energy features; this is often of greatest qualitative interest when an experimentalist seeks an interpretation of a hopelessly dense spectrum of innumerable lines. Quantum chaos techniques interpret the main global features in terms of a modest number of periodic classical orbits. The new capabilities of such theories are deserving of special scrutiny from this scientific community. Their full domain of applicability remains to be identified. It is encouraging that for a class of problems involving Rydberg state dynamics in external fields, they have provided unparalleled insights into the origin of the short-time spectrum.

For decades, applied mathematics has been exploring problems relating to the asymptotic distribution of eigenvalues, a classical area in mathematics that has been reinvigorated by periodic orbit theory. More generally, theoretical atomic physics would also

benefit from stronger interaction with the larger applied mathematics community. That community contains expertise in areas of key relevance to atomic physics, including differential equations, numerical methods, and asymptotic analysis.

Low-Temperature Plasma-Related Research

The problems presented in understanding the physics of high and low temperature plasmas lie at the forefront of atomic, molecular and optical physics. The behavior of plasmas is determined by a complex of interactions of a wide array of atomic, molecular and optical processes involving electrons, ions, atoms and molecules. The full characterization of many of the processes that determine the nature of plasmas wait upon advances in theoretical concepts and developments in computational methods. Thermal fusion plasmas, inertial confinement plasmas and plasmas designed to simulate weapons effects are hot, with temperatures approaching one million to ten million degrees. Similar hot plasmas occur naturally in the solar corona, in supernova remnants and in gas surrounding cosmic X-ray sources. The behavior of high temperature plasmas is determined by ionization, recombination and excitation processes. The efficiencies of many of the processes can be calculated by the application of methods that have been already developed and tested. For others, and particularly for systems of high nuclear charges, further development is needed that takes fully into account relativistic effects. Using the data, computer-based models that simulate the behavior of the plasmas can be constructed. The plasma properties can then be explored over a wide range of conditions of density, temperature and radiation environments and diagnostic probes can be designed.

Lower temperature plasmas are created for manufacturing purposes in the lighting and plasma processing industries. They occur naturally in the ionospheres of the planets, in lightning discharges in the terrestrial atmosphere and in the interstellar medium. The behavior of cooler plasmas with temperatures below about 5000 K is complicated by the presence of neutral and ionic molecules and negative ions. Dust may be formed and will exercise a major influence on the characteristics of the plasmas. The molecular composition is drastically modified by the production of metastable species and by the vibrational excitation of the molecular species. Important processes are dissociative recombination, dissociative attachment, associative detachment, collisional dissociation, and chemical reactions involving excited species. Few data are available and for many of the processes no adequate theory has been formulated. The processes of dissociative recombination (DR) of molecular ions with electrons and charge transfer reactions of ions with molecules, in particular, pose formidable theoretical challenges. For even some of the simplest diatomics (HeH^+) and triatomics (H_3^+), theory and experiment disagree about the near-threshold DR cross sections by more than an order of magnitude.

Present models of plasmas contain only a limited description of the processes and are based on unreliable data. With anticipated advances in collision theory for complex systems and access to enhanced computing capabilities, it will be possible to identify the critical processes, to calculate their efficiencies and to construct models that can be used to guide the design of low temperature plasmas for specific applications.

Ion-Atom and Ion-Molecule Collisions

New data are becoming available for many different kinds of reactions. As the possibilities of observing transitions and reactions in many-electron systems grow, so do the difficulties of finding which studies will provide the best insight into the nature of dynamical processes in complex systems. In these cases theory can often provide guidance in selecting reaction details most crucial to understanding mechanisms for dynamics in complex systems. A challenge in the field of collisions is to unravel reaction mechanisms among many-electron atoms, molecules, clusters, and surfaces, including instances in which perturbation techniques fail. In this field

experiments carried out at ion storage rings have provided critical data at unprecedented resolution for a number of electron-ion recombination and collision processes. This is an area in which the United States has fallen significantly behind development in Europe. Smaller, less expensive, "table-top" storage rings, which should be available soon, provide an important opportunity for U.S. science to explore many of these topics.

OPPORTUNITIES AND HORIZONS IN HIGH-PERFORMANCE COMPUTING

Progress in this century in atomic, molecular and optical theory has placed the field in an enviable position to make accurate predictions about fundamental processes. However, for the complex systems and processes of increasing relevance to this field, the impact of theory on experiment can be increased dramatically through use of the most powerful modern computers and computing techniques. The significant increase in performance provided by such computers will enable AMO theory to address fundamental processes in more complex systems. Furthermore, as many branches of AMO theory make the transition to parallel computing, within a decade the applications developed for today's massively parallel computers will be running on widely-available laboratory-scale multiprocessors.

The accurate theoretical description of collisions among electrons, ions, atoms, molecules, clusters and surfaces cannot be achieved without this scale of computation. Such advances will allow us to predict the rates of the many important collision processes and to construct simulations of the low-temperature plasmas used widely in processing materials. They will also permit studies of bound and continuum states of heavy elements and of new phenomena occurring in complex systems in intense fields. A further impact of high-performance computing is the opportunity to make the computational tools of modern theory more accessible to the experimental community by moving beyond the "grand challenge" stage to the point where use of such tools pervades the entire AMO community. Many AMO codes (RMATRIX, MCHF, GRASP, and Cowan's suite of programs) have been developed and used by theory groups, but little attention has been given to making them user-friendly and readily accessible to experimentalists. Other communities such as the chemistry community have begun to make this transition. Here, quantum chemistry codes such as GAUSSIAN, GAMES, QCHEM and others have been become the every day tools of organic and inorganic chemistry. Few similarly-accessible tools exist for the interpretation of experimental results for a wide range of problems that involve dynamics, such as electron collisions, photoionization, and molecular and ionic collisions. With the advent of new DOE national experimental facilities such as the Advanced Light Source and the Advanced Photon Source, the need for such tools and for closing the gap between theory and experiment is more urgent than ever.

Panel E: Nano- and Mesoscopic Structures

Chair:
Paul Alivisatos (Lawrence Berkeley National Laboratory)

Panelists:
Daniel Chemla (Lawrence Berkeley National Laboratory)
Vicki Colvin (Rice University)
Robert Compton (University of Tennessee)
Therese Cotton (Iowa State University)
James Heath (University of California, Los Angeles)
Mark Knickelbein (Argonne National Laboratory)
Charles Lieber (Harvard University)
Paul McEuen (University of California, Berkeley)
Horst Schmidt-Boecking (Universitat Frankfurt)
Stanley Williams (Hewlett-Packard Laboratories)

Solids of nanometer scale exhibit strongly size dependent chemical and physical properties which represent limiting behaviors for different types of matter (atomic to bulk). The variations with size are enormous, and this represents a new opportunity to optimize material properties by varying their size and shape rather than by changing their chemical composition. The developments in nanoscale science are of a sufficiently fundamental nature that we can anticipate the development of this field will significantly impact several other disciplines of basic science and will also help address key energy related technologies.

Advantages and Challenges of Nanometer Scale Science and Technology

The ability to shape and control matter on the nanometer scale offers a tremendous number of established and potential advantages, but to fully exploit this area there are a number of critical issues that must be addressed over the long term. In this section of the report, we discuss both the major advantages that nano-scale based systems offer to new and/or improved technologies, as well as the major questions that surround the development of any such new technologies.

The primary advantage of any nanostructured material lies in the extensive tunability of its properties. For example, the physical and chemical characteristics of materials have traditionally been determined by the chemical stoichiometry and structure of the material. However, by controlling the size and shape of a nano-scale solid, many of the physical and chemical properties of the system can be tuned over a large range. For example, the fundamental characteristics of a material, such as its melting temperature, color, saturation magnetization and coercivity, charging energy, chemical reactivity, etc., are all a function of size and shape. For instance, the color of semiconductor quantum dots can be varied continuously from the near infrared to the ultraviolet. Such color changes correlate to electron and hole energy levels, which in turn affect the catalytic and chemical behavior of the particles. Thus, nano-scale building blocks lend major new experimentally controllable variables for fabricating desired materials. In the last decade, it has been possible to fabricate nanostructured materials at a level where they now compare in quality to the high performance solids and thin films used in the electronics industry. A partial list includes highly monodisperse colloidal metal and semiconductor nanocrystals, STM-assembled "quantum corrals," molecular beam epitaxy grown self-assembled quantum dots, laser ablation and cluster beam expansions, electric discharge growth of carbon tubes and clusters, thermal flow reactors, rapid pyrolysis, as well of course as e-beam and x-ray lithography. Atom manipulation, and matter diffraction from light waves, are important new tools emerging from atomic and optical physics which may lead to new ways of fabricating nanostructures.

Despite the wide range of fabrication methods, a high degree of structural perfection is often observed in nanostructures, and this can be attributed to several factors: In a bulk single crystal, it is impossible to remove all defects. In a nanostructure, the high surface area provides a route for the annealing and removal of defective sites on short time scales. Related to this is the fact that the melting temperature of solids is substantially reduced in finite size, which allows not only for the fast diffusion of defects, but also for growth schemes at surprisingly low temperatures. Thermodynamically, the probability of defect formation dictates the fraction of atoms likely to be in defect sites; when the total number of atoms in the system is small, this number approaches zero. These arguments suggest that the manufacture of nanostructures can be done very cheaply, with simple processing. This makes nanostructures extremely attractive candidates for a host of technologies.

As the quality and variety of nanostructures has improved over the last decade, many new technological applications have been envisioned. This, in turn, has raised several questions related to the integration and performance of nanostructures in

various technologies. First, can sufficient quantities of high uniformity nanostructures be made to enable technological applications? All current fabrication schemes operate on a typical laboratory scale (ranging from 1 monolayer to 1 gram). Second, one of the advantages of nanostructures is that they are characterized by a very high surface area. However, this also makes interface characterization and control imperative. Opportunities for the development of new spectroscopic and imaging tools exist here. There is as yet no tool available to determine the bond lengths, angles and reconstructions of nanoparticles, as exists for plane single crystal surfaces. Third, many envisioned applications require nanostructures to be placed into chemically and/or structurally complex environments – i.e. around other nanostructures, connected to electrical devices, in chemical sensing environments, etc. The assembly of nanostructures into specific architectures is a challenge that is just now beginning to be addressed. Fourth, the chemical and thermodynamic stability requirements for nanostructures will vary from application to application. This issue remains largely unexplored.

Nanostructures are extremely promising new materials, with significance for both basic and applied science. Although the field of nanostructures is young, it is already possible to identify key areas in which nanostructure science must evolve for the field to reach its full potential. At present time, none of the questions discussed here appear insurmountable.

Impact of Nanoscience on Fundamental Problems

Nanostructure studies are most often described in terms of their potential technological impact, to the point where it is a field often referred to as nanotechnology. Yet the ability to control matter on the nm length scale can be expected to have important consequences for fundamental studies in disciplines ranging from condensed matter theory, to solid state chemistry, to materials science.

An isolated nanometer scale object such as a nanocrystal or a nanotube can be viewed as an artificial atom, possessing a well-defined charge state and energy level structure. Unlike real atoms, however, the size, shape, properties, and local environment can be controlled and adjusted. As a result, nanostructures are ideal model systems for exploring the physics of small quantum systems.

The electronic structure of the delocalized electrons in a nanostructure can be continuously tuned by adjusting the size of the object. This shifts the absorption and emission spectrum from the bulk material and results in discrete lines. Further, the charge state of the nanostructure can be tuned using a voltage applied to an external electrode, continuously adjusting the ionization energy of the artificial atom. Finally, the shape can also be adjusted. For example, 3D spherical nanocrystals, 2D disc shaped quantum dots, and 1D rod-like molecules are currently under study to investigate the energy level structure of systems of different dimensionality. This tunability makes possible the systematic investigation of the relative importance of Coulomb interactions and quantum confinement effects in small quantum systems. It also makes possible the creation of artificial quantum systems with properties that are specially chosen for a specific scientific application. This ability to control the electronic structure is of considerable interest from the point of view of interaction of light with matter and quantum optics. The non-linear optical properties and the question of electronic coherence in dot excited states are particularly related to developments in atomic and optical physics. For example, dots are possible candidates as qu-bits in quantum computation.

The artificial atoms can also be assembled into artificial molecules and solids, and the properties of this solid can be controlled. In particular, the level structure of the individual nanostructures, their charging energy, their Coulombic and tunnel coupling, and their coordination can all be tuned. This is tremendously important for understanding the

behavior of interacting electrons in reduced dimensions, which is probably the central task facing condensed matter physics. From high temperature superconductors to Luttinger liquids, the non-Fermi liquid ground states that are believed to occur in 2D and 1D interacting electron gases remain poorly understood, both due to a lack of tunable experimental systems and their intrinsic theoretical difficulty. The optical, electrical, and magnetic properties of such structures will be of great interest.

Nanoscale materials provide many opportunities for improved understanding of basic chemical processes. The study of chemical reactions occurring on and within nanoscopic objects such as clusters, dots and nanotubes provide a unique opportunity for examining the effects of reduced dimensionality. One example with environmental implications involves the study of chemical reactions of atmospheric pollutants within water nanodroplets and on the surface of ice nanocrystals. A second example concerns our understanding of solid state chemistry. Solid state synthesis is generally thermodynamic, largely because mechanistic thinking is difficult to implement in inhomogeneous solids with defects present. Nanostructures provide the opportunity to prepare pre-assembled, multi-component single grains, in which the kinetics and mechanisms of solid state processes can be independently measured and controlled. The studies of metal cluster nanosurfaces provide an opportunity to examine intrinsic (electronic of geometric) size effects on surface reactions of catalytic importance such as Fischer-Tropsch synthesis and hydrodesulfurization. The newly developed ability to put stable molecules as well as molecular coreactants in finite cluster solvents, and in liquid helium nanodroplets opens the possibility performing spectroscopic and reactivity studies at temperatures below 1K.

Nanometer scale materials such as semiconductor nanocrystals and carbon nanotubes can be created with no defects. This allows for fundamental materials science studies of the properties of pure materials, e.g. the strength or structural metastability. In bulk systems, these properties can be influenced by defects. The ability to create perfect materials makes measurements of the intrinsic, i.e. defect free, properties possible. The mechanical properties of ultrasmall structures are also of fundamental interest, e.g. size quantization of phonon modes and quantum "mechanical" oscillators. A new field of nanomechanics is emerging that explores the physics and technology of mechanical structures at this scale.

The Role of Theory

Theory plays a central role in the description of the structure and dynamics of nanostructures as well as to predict the growth of such materials, and can help to guide experiment. In turn, nanostructures provide an important proving ground for new theoretical developments. The ability to describe complex systems is of particular theoretical interest, and the evolution with size of nanostructure properties provides a heirarchy of complexity.

The theoretical description of isolated nanostructures is just beginning. Vastly different computational methods such as ab initio quantum chemical calculations, density functional theory coupled with molecular dynamics, and simple tight binding approximations all yield electronic ground and excited state energy levels for neutral and charged isolated nanostructures. In some cases the applications of semi-empirical methods combined with experimental benchmarks can be used to generate useful predictions of a wide range of related structures such as functionalized nanoparticles. The issue of predicting equlibrium structures, including geometry optimization and surface reconstruction including the presence of ligands or other interface atoms is particularly important. A number of theoretical methods can be applied to the mechanisms and kinetics of nanostructure growth. The calculation of vibrational and electronic density of states as well as polarizability and multipole moments can be compared with experiment. Of particular importance is the prediction of magnetic properties of nanostructures.

The effects of geometry changes and external stress on these properties is of interest.

The physical and chemical mechanisms whereby nanostructures, for example quantum dots, are coupled to the environment and to each other are of great interest. Properties such as electron transfer rates between quantum dots, artificial solid band structures and collective excitations in coupled dots need to be addressed. There are many questions regarding the interactions of dots coupled to the environment. Empirical potentials for dot-dot and molecule-dot interactions together with the long range forces responsible for the assembly of dot arrays are importance. Of equal importance is the chemical reactions occurring at dot surfaces.

Relationship to Energy Technologies

Almost every basic energy related technology may be impacted by developments in nanoscale science. These include: chemical technology, energy storage and manipulation, information storage and manipulation, and structural and mechanical materials.

Catalysis inherently relies on nanoscale components and can be expected to benefit greatly from the ability to rationally control the size and shape of components with precision and to spatially organize the components. Nanostructures are also exquisitely sensitive to changes in their chemical and physical environment, making them strong candidates for sensor elements; for this to be realized issues of selectivity and amplification these must be addressed. Nanostructures also provide a model system for studying reactions on aerosols, which in turn are important for understanding atmospheric chemistry and pollution remediation.

Photovoltaics, light-emitting diodes, batteries, fuel cells and capacitors are some of the *energy storage and manipulation* technologies that may benefit from the high surface to volume ratios provided by nanostructures. Nanostructures have the desirable physical properties of solid systems with the added benefit of the ease of processing of molecules. Composites of inorganic nanoparticles with organic polymers are of particular interest in this category, and every type of device mentioned above has been demonstrated in composites

A bit is a physical entity, and there is an energy cost associated with *information storage and manipulation*. In the past 25 years the energy efficiency and consequently the speed of computation per input power have increased by a factor of ten thousand. Silicon integrated circuit technology should be able to deliver another factor of one hundred increase before it reaches technological limits. However fundamental physical limits indicate that computers can improve another factor of ten billion in energy efficiency. To approach the thermodynamic limit it will be necessary to construct computers from nanostructures and several possible models for subsystems already exist. Computing will benefit tremendously from the combination of decreased energy consumption, increased speed, and enormous storage increases (x 10^{10}) that will be enabled by nanostructures.

Magnetism is fundamentally different in nanosize grains, which act as single domains. If the shape and arrangement of magnetic nanostructures can be controlled, this will have uses in magnetic storage and sensors.

Nanoscale building blocks tend to spontaneously exclude defects. As a consequence, the *structural and mechanical properties* of the individual components and of their assemblies are quite distinct from those of extended solids. The striking example of this is carbon nanotubes which appear to be the strongest fiber ever discovered. Nanostructure composites with well defined architectures will have unusual and desirable mechanical properties.

Relationship of AMO Physics to Nanoscience

Atomic, molecular and optical physics has contributed greatly to the development of the emerging field of nanoscience. Nanostructures and clusters, in fact,

represent a "bridge" between the isolated atom or molecule to the condensed matter state. Following the development of nozzle-jet expansions for the production of gas phase clusters, many AMO scientists have contributed to our understanding of such diverse phenomena as solvation, the hydrated electron, chemistry of nanosurfaces, etc. More recently, the assembly of nanostructure materials at the macroscopic level has produced a heightened level of interest in the AMO community. Many of the traditional techniques inherent in AMO physics are being directly applied to the study of nanostructures, e.g. mass spectroscopy, electron spectroscopies, photon spectroscopy, etc. In fact, the discovery of C_{60} came about through the combination of a number of these techniques: (1) laser ablation of graphite into a nozzle jet expansion, (2) laser multiphoton ionization of C_{60} followed by (3) time-of-flight mass spectroscopy. In addition to these standard techniques, there are a number of new advances in AMOP techniques which might be important to the development of nanostructure science:

Many possibilities exist for transferring experimental techniques from atomic physics to nanoscience. Recent developments of laser cooling of atoms might be applied to the partial cooling of nanostructures. The ability to cool these clusters would lead to a better interpretation of their spectroscopy. It is also conceivable that such cooling could lead to new molecular states of matter.

The study of the ionic properties of nanostructures represents an exciting new field for AMO physics. Many of the ionization processes operative in nanostructures do not occur on the atomic or molecular level. For example, the ionization of clusters such as C_{60} occurs through surface plasmon excitations. The production of new "super atoms" such as metal endohedral fullerenes will afford studies of energy levels of interest to solid-state heterostructures. The attachment of electrons to nanostructures represents a new field of physics in which techniques of AMOP are essential. Nanostructures provide templates for the first detailed studies of multiply charged negative ions.

Many developments in optical physics may impact nanoscience. As one example, the "holy grail" of laser chemistry is to coherently control the products of reactions by the application of shaped optical pulse sequences. This is a very active research area. It is possible that advances in this field could contribute to the "assembly" of nanostructured materials, or to the manipulation of electronic coherence in quantum dot molecules and solids. The interaction of intense optical fields with nanostructures is of interest. Multiphoton ionization, and the measurement of nonlinear susceptibilities as a function of the size are currently actively investigated. The ability to control size may provide more insight into the differences between atoms and molecules in their non-linear responses.

It is possible to reduce the size of electrically controlled macroscopic objects to sufficiently small dimensions that the nanostructure can be manipulated by the application of an external electrical field, e.g. a very high field applied between two neighboring quantum dots can directly influence the reactions of these quantum dots with their neighbors or scattered projectiles. In such strong fields, collisions of atoms or molecules with the nanostructures might allow electron transfer from the quantum dot to the colliding atom or tunnel directly to vacuum states. Such enhanced electron emission processes might find applications in, for example, electron-induced catalysis, air pollution abatement, etc. By varying the size, geometry, and external field one may also obtain direct information regarding the energy levels of nanostructures.

Traditional electron, ion, atom, etc. beams can be used to excite or even fragment these nanostructures to determine their structural and dynamical properties. In addition, new atomic beam microscopies (e.g. cold supersonic He beams or atom laser beams) could be used to study the topography and phonon structure of

the surface. Using seeded supersonic cluster beams, or possibly laser cooling, very cold nanostructures may be created.

Cold atomic beams can create interference patterns of atoms with order 100 nanometer resolution. In this case, the AMOP techniques for producing such cold beams are directly applicable to lithography. Furthermore, using UV or even low energy x-rays from synchrotron light sources, 3-dimensional nanostructures or layers can be produced.