

Summary of Research Presentations at the CAMOS Meeting in April 2012

Coherent Control of Atomic and Molecular Processes

The National Research Council's (NRC's) Committee on Atomic, Molecular, and Optical Sciences (CAMOS) met on April 3-4, 2012 at the Keck Center in Washington, D.C. A portion of this meeting focused on recent advances in coherent control of atomic, molecular, and optical processes. The topic was addressed in one of the six scientific chapters of the NRC's 2007 report *Controlling the Quantum World: The Science of Atoms, Molecules, and Photons*, known in the AMO field as the 2010 AMO Decadal Survey. Five speakers presented talks that surveyed a sampling of current research in coherent control: Herschel Rabitz (Princeton University), Marcos Dantus (Michigan State University), Tamar Seideman (Northwestern University), Thomas Weinacht (State University of New York in Stony Brook), and Mikhail Lukin (Harvard University). The topics discussed included general considerations about the method of optimal control and its limitations (Rabitz); applications of phase control in laser-matter interactions (Dantus); control of rotational and torsional motion in isolated molecules and in solution (Seideman); strong-field control of molecular dynamics (Weinacht); and control of nitrogen vacancies in diamond as a solid-state, atom-like hybrid system (Lukin).

To some extent, the goals of laser control in chemistry and AMO physics have evolved over the past decades. In particular, the use of lasers for bond-selective chemistry as a practical method for high-yield synthesis is now much less a focus than the use of laser control to enhance sample preparation and measurement technologies, as well as to provide insight into the mechanisms of previously opaque processes in chemical dynamics and reactivity. In many ways, the applications of quantum control have become far broader and more pervasive than originally expected, as illustrated by the broad range of topics in the focus session.

SPECIFIC OBSERVATIONS

General Implications of Optimal Control Theory in Chemistry

From a general perspective, there are two potential goals for quantum control: to achieve an objective and to understand system properties and reaction mechanisms. The use of adaptive feedback control in laser chemistry experiments has proven to be quite powerful, and the approach can be

applied to many more general problems—for example, the development of catalysts for selective oxidation reactions. One of the surprising features of this approach is that optimal solutions can often be found by sampling a relatively small fraction of the entire search space. For example, optimal solutions in laser control experiments could be found with only 10^3 - 10^4 iterations in a problem with a search space of dimension 10^{20} or higher, raising the question of whether this high search efficiency is a coincidence. This question has led to an examination of the nature of the control landscape—that is, the dependence of the targets of interest on the control variables. With a small number of assumptions, the high search efficiency emerges as a property of the control landscape. How widely applicable these assumptions are remains open to debate. Investigations into the general utility and limitations of optimal control techniques can provide insight into very general issues about problem solving and are expected to continue to be an active research area.

Applications of the Phase Control of Laser–Matter Interactions

Automated approaches based on optimal control techniques have made the production of ultrafast optical pulses with tailored frequency and phase distributions almost routine. This capability has enormous potential not only for opening new avenues for fundamental research but also for creating a broad range of practical applications. For example, two-photon excitation with tailored laser pulses can be used for selective electronic and vibrational excitation in complex systems, allowing the label-free, chemically specific imaging of living cells by stimulated Raman scattering microscopy. Controllable ultrafast pulses have been shown to allow selective bond breaking in complex proteins, which may ultimately allow simplified methods for the analysis of protein structure. Tailored ultrafast laser pulses can be used to optimize the damage created by laser ablation techniques, leading to higher resolution laser-created structures in materials. Many more applications were also discussed, and it appears very likely that the routine ability to create tailored ultrafast laser pulses will find many new, as yet unexplored applications in the coming years.

Strong Field Control of Molecular Dynamics and Control-Based Spectroscopy

Work on the strong-field control of molecular ionization and dissociation using shaped ultrafast optical pulses has considerably improved our understanding of ionization and dissociation in intense laser fields. Such studies use closed-loop, feedback approaches with well-defined targets (e.g., ionization yield and product branching ratios) to determine optimal laser pulses. Subsequent analysis of the pulse shapes, including both the phase and intensity distributions, can be compared with detailed theoretical calculations of the relevant potential energy surface(s) and dynamics to allow the

reconstruction of the ionization or dissociation dynamics and thereby an understanding of why a particular pulse shape is optimum. Laser control can also be used to produce selective stimulated emission from particular species even in the presence of overlapping absorption and emission features from other species in the sample. This approach has been demonstrated both in atomic vapors and for complex molecular systems in solution. The development of similar control schemes to isolate spectroscopic signals of interest from complex samples should find considerable utility in a wide range of applications.

Coherent Alignment in Complex Systems

Over the last two decades, methods have been developed to control the rotational degrees of freedom of molecules by using moderately intense laser fields, allowing the preparation of aligned ensembles of molecules that can be used in a variety of dynamical studies. Specifically, these quantum control techniques can be used to prepare samples that greatly simplify data analysis and dramatically enhance the ability to extract important information from the data. While these techniques were originally developed for rigid, isolated diatomic molecules, methods have been developed that allow the three-dimensional alignment of arbitrary molecules, and new methods have recently been proposed to allow the alignment of molecules in solution. As an illustration of the power of alignment techniques, the ability to align gas-phase samples of large biomolecules is expected to be very important for the analysis of x-ray imaging studies at free-electron laser facilities such as the Linac Coherent Light Source. As another example, alignment techniques have been used to enhance high-harmonic generation in strong-field studies of small molecules. Aligned molecules are also expected to have considerable utility in collision studies, where the relative orientation of collision partners is expected to have a substantial effect on energy transfer processes. Recent work has also shown that it is possible to control the torsional motion of larger molecules. Moreover, because the electronic behavior of such systems can depend strongly on torsional degrees of freedom, this capability may have a number of interesting applications. Finally, the ability to control molecular alignment may ultimately allow the detailed control of molecular assembly. Indeed, this approach has already been used to guide the orientation of molecules attaching to surfaces.

Control of Nitrogen Vacancies in Diamond

Nitrogen vacancy (NV) impurities in diamond are "nature's trapped ion," providing a robust solid-state atom-like system that can be manipulated and controlled. Many potential applications for this system are being explored, and considerable progress has already been made in a number of areas.

Controlling the isolated electronic and nuclear spins in NV centers can provide a solid-state qubit memory, and approaches for building scalable quantum computing architectures have been proposed. Isolated NV centers in diamond can also provide bright and stable single-photon sources, and these have been used to demonstrate interference and the potential for entanglement of photons from separate remote sources. The spin on the NV center can also be used as a sensor that can be coupled with nanomechanical oscillators to provide new probes of quantum-level effects. NV centers in diamond nanocrystals have also been used as a probe of nanoscale processes in biological systems, both as a fluorescence marker and as a sensitive magnetometer to probe flow through ion channels in cell membranes. While there is a considerable effort to develop synthetic methods to produce artificial NV diamond by N ion implantation techniques, for many applications natural NV centers still provide superior performance. This situation will likely improve as new applications continue to be developed and larger scale integrated structures are explored.

GENERAL DISCUSSION

In the general discussion following the session, the broad range of applications for quantum control was noted, as well as the interdisciplinary nature of much work in the field. While the original goal of controlling arbitrary chemical reactions to efficiently produce high yields of desired products remains in the distance, quantum control techniques have become powerful tools in a broad range of applications and are a fundamental part of many forefront areas of AMO physics. Indeed, the key role of quantum control techniques is reflected in the title of the AMO 2010 Survey, "Controlling the Quantum World." Quantum control techniques are expected to become even more important in the coming years.