The 2011 Jerome B. Cohen Distinguished Lecture Series

Dr. Emily Carter
Department of Mechanical and Aerospace Engineering,
Program in Applied and Computational Mathematics,
and Gerhard R. Andlinger Center for Energy and the Environment
Princeton University

Contributing to Efficient Power Generation: Atomic-scale Insight into Thermal Barrier Coatings from Quantum Mechanics

Wednesday, June 1, 2011 **4:00 p.m.** **Tech, Lecture Room 211**

To maximize energy efficiency, gas turbine engines used in airplanes and for power generation operate at very high temperatures, even above the melting point of the metal alloys from which they are comprised. This feat is accomplished in part via the deposition of a multilayer, multicomponent thermal barrier coating (TBC), which lasts up to approximately 40,000 hours before failing. The TBC consists of a NiAl-based bond coat alloy layer onto which yttria-stabilized zirconia (YSZ) is deposited. A layer of alumina (or "thermally grown oxide," TGO) grows in between these two materials during YSZ deposition, which then subsequently thickens during engine use. To extend engine service lifetime or to increase the gas inlet temperature (thereby increasing efficiency), it is critical to understand mechanisms of failure and then to design circumvention strategies. We use first principles quantum mechanics - specifically periodic density functional theory (DFT) - calculations to test hypotheses about impurities that harm TBCs and transition metal (TM) dopants that extend TBC lifetime, including the role of Pt and early TMs in NiAl-based bond coat alloys. For example, it has been shown experimentally that TGO thickening is correlated with TBC failure. Thus, hindering alumina growth is thought to be critical to prolonging TBC lifetimes. Empirically, it is known that TMs such as Pt, Hf, and Y improve the stability of the TBC, though their mechanism of action is not well characterized. Some of these TMs (e.g., Y) segregate to grain boundaries in the TGO, and it has been suggested that alumina growth may be inhibited by their presence. As it is thought that growth of the alumina layer is controlled by Al and O diffusion at grain boundaries, we explore the structure and energy landscape at an alumina grain boundary for Al, O, early TMs, and rare earth elements. We also consider how Pt affects the thermodynamic stability of defects and high temperature diffusion kinetics in the NiAl bond coat, in order to elucidate the role of Pt in stabilizing TBCs. Implications for growth of the alumina layer and the fate of TBCs, as well as proposed new dopant to improve TBC robustness and ultimately energy conversion efficiency, will be discussed.

Professor Carter is the Founding Director of the Andlinger Center for Energy and the Environment at Princeton University, the Gerhard R. Andlinger Professor in Energy and the Environment, and Professor of Mechanical and Aerospace Engineering and Applied and Computational Mathematics. She earned her B.S. (Chemistry) from UC Berkeley in 1982 and a Ph.D. (Chemistry) from Caltech in 1987, spent a brief postdoc at CU Boulder, and then was on the UCLA faculty for 16 years in Chemistry and later also Materials Science and Engineering. She moved to Princeton in 2004, where she is also associated with Chemistry, Chemical Engineering, and three interdisciplinary institutes. The author of over 240 publications, she has delivered over 400 invited lectures worldwide. Her scholarly work has been recognized by numerous honors, including election in 2008 to both the American Academy of Arts and Sciences and the National Academy of Sciences, and in 2009 to the International Academy of Quantum Molecular Science. Her current research is focused entirely on enabling discovery and design of materials for sustainable energy, including converting sunlight to electricity and fuels, providing clean electricity from solid oxide fuel cells, clean and efficient combustion of biofuels, and optimizing lightweight metal alloys for fuel-efficient vehicles.

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Quantum Mechanical Evaluation of Energy Conversion Materials for Generating Electricity and Fuels

Thursday, June 2, 2011 **4:00 p.m.** **Tech, Lecture Room 211**

We have launched three major research efforts to use quantum mechanics techniques to search for robust, efficient, and inexpensive materials for solid oxide fuel cells (SOFCs) that convert fuels to electricity, photovoltaics (PVs) that convert sunlight to electricity, and photo-catalytic electrodes (PCEs) that convert sunlight, carbon dioxide, and water into fuels. Various observables that are key metrics for determining the utility of a given material can be accurately calculated from quantum mechanics; we will discuss our theoretical schemes for each observable and how we validate our approach. In our SOFC research, we are focusing on cathode optimization, often considered the limiting factor in reducing the high operating temperatures of current SOFCs. Porous electrodes can be readily synthesized for SOFCs such that gas transport is facile. If oxide ion diffusion and electron transport could be enhanced, along with rapid dissociative adsorption of dioxygen on the cathode surface, lower temperatures could be used, which would facilitate wider deployment. In the solar energy conversion arena, the cost-efficiency tradeoff for PV materials motivates a look at new options and despite periodic media reports to the contrary, no efficient PCEs are available yet. I will discuss why it is so difficult to find effective PCE materials; in particular I will enumerate the very significant constraints beyond those on PVs that they must satisfy to achieve high efficiency. Limiting oneself to abundant elements further constrains the design space. As a result, we are focusing primarily on first row transition metal oxide materials. Key properties of conventional and novel materials, along with some new design principles, will be discussed. The work is revealing which dopants or mixed oxides are likely to provide the most efficient energy conversion materials.

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Quantum Simulations of Materials at the Mesoscale: Physics, Algorithms, and Applications

Friday, June 3, 2011 **4:00 p.m.** **Tech, Lecture Room 211**

Many materials phenomena are controlled by features at the mesoscale, *i.e.*, a length scale above atoms but below the micron scale. In addition to the fashionable example of nanostructures, other practical examples abound. For instance, the mechanical properties of metals are largely controlled by the nucleation and motion of dislocations and their interaction with other defects (*e.g.*, grain boundaries, solutes, precipitates) in crystals. Experiments (*e.g.*, electron microscopy) provide post-mortem examination of these features. By contrast, computer simulations can interrogate these defects *in situ*. Of course, reliability of the simulations is always an issue. Our research aims to develop predictive simulation tools that do not rely on any experimental input, such that they produce a truly independent source of data for comparison with experiment. This assumption/empirical-input-free approach requires going back to basic physical laws, namely those of quantum mechanics to describe electron distributions in materials. Normally such techniques are prohibitively expensive for simulating more than a few hundred atoms on supercomputers. But because of our recent algorithmic improvements to a quantum mechanics method (orbital-free density functional theory) that makes it scale fully linearly with system size, we are now able to simulate fully quantum mechanically and accurately large scale defects that play key roles in plastic deformation and ductile fracture of main group metals and metal alloys, with accuracy rivaling the most accurate solid state quantum mechanics methods available. Moreover, we will discuss a breakthrough in the description of the kinetic energy of electrons (via a new kinetic energy density functional) that extends the regime of reliability of this technique beyond main group metals to semiconductor materials. Our current applications are focused on examining the behavior of dislocations in aluminum and magnesium; ultimately we hope to optimize the composition and microstructure of lightweight metal alloys (by finding the sweet spot in the ductility-strength tradeoff) that can be used to improve the fuel efficiency of vehicles.

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