Wednesday Morning, October 30, 2013

Accelerating Materials Discovery for Global Competitiveness Focus Topic
Room: 202 B - Session MG+EM+MI+MS-WeM

Materials Discovery and Optimization through Iterative Approaches

Moderator: M.B. Holcomb, West Virginia University, S. Jones, National Science Foundation

8:40am **MG+EM+MI+MS-WeM3 Preparation of Ultra Stable Organic Glasses by Physical Vapor Deposition**, ***J.J. de Pablo***, University of Chicago, *L. Yu*, *M.D. Ediger*, University of Wisconsin-Madison **INVITED**

There is considerable interest in identifying structure-property relations in glasses. Structural studies of glassy materials have benefited from insights provided by molecular simulations of model glass forming liquids. In particular, simulations have provided support for the existence of dynamic and mechanical heterogeneity at the level of small groups of molecules or particles. In general, however, the cooling rates employed in simulations have been many orders of magnitude faster than in experiments, thereby adding some level of ambiguity to direct comparisons between theory and experiment. Recently, experiments have shown that glasses of organic molecules having unusually large thermal and kinetic stability can be prepared by a vapor deposition process. Such glasses exhibit higher onset temperatures, higher densities, and higher mechanical constants than those of ordinary glasses. In particular, vapor-deposited glasses have been shown to exhibit relaxation times that are many orders of magnitude longer than those of ordinary glasses. Inspired by such experiments, we have devised a strategy that allows one to prepare highly stable glassy materials, in silico, having thermal and kinetic characteristics that are superior to those obtained by gradual cooling of liquid samples. In this presentation, we will describe current experimental efforts to produce and characterize stable glasses, along with an overview of available theoretical and computational strategies aimed at understanding their properties. Emphasis will be placed on recent attempts to identify the structural origins behind the extraordinary stability of vapor deposited glasses, with the goal of providing new insights for design of stable amorphous materials deep in the potential energy landscape.

9:20am **MG+EM+MI+MS-WeM5 Composition and Structure Manipulation for Energy Materials with Improved Properties**, ***S. Lany***, *H. Peng*, National Renewable Energy Laboratory, *V. Stevanovic*, Colorado School of Mines

Ideally, materials for energy applications like photovoltaics, photo-electro catalysis, or thermoelectrics should have a high performance, but be earth-abundant and manufacturable by low-cost processes. Given such demanding constraints, the list of promising materials that could potentially fulfill all requirements often shrinks to a few hopefuls. Then, the question arises whether we can start from materials that have both good and bad features, and manipulate the composition and structure such to cure their deficiencies. The role of theory is to assess the properties and identify promising alloying approaches.

We are presenting two recent examples for this approach: As a photovoltaic material, Cu2O suffers from a high absorption threshold due to a relatively large band gap and a forbidden optical transition. Also, the p-type doping is 1-2 orders of magnitude below the optimal level. Our theoretical work identifies alloying of Zn, S and Se as a promising route to improve the optical and electrical properties of Cu2O. Strong dopant-defect interactions lead to the unusual situation that the isovalent (S, Se) alloying increases the electrical doping, whereas the aliovalent (Zn) alloying changes the optical properties. The second example of MnO-ZnO alloys is based on the prediction [Phys. Rev. B 85, 201202(R) (2012)] that MnO in a hypothetical tetrahedral structure (zinc-blende or wurtzite) should have a smaller band gap and superior carrier transport properties than the normal octahedral rocksalt structure. These features would make this hypothetical material interesting, e.g., for photo-electro-catalytic water-splitting. We predict that $Mn_{1-x}Zn_xO$ alloys assume the wurtzite structure above x = 0.38, and that such alloys preserve the beneficial properties of the tetrahedral MnO phase. Thus, the alloy approach presents a viable path to realize desirable materials properties that were originally identified for a hypothetical material.

9:40am **MG+EM+MI+MS-WeM6 Accelerated Optimization of Solar Cell Materials**, ***A. Zakutayev***, National Renewable Energy Laboratory, *V. Stevanovic*, Colorado School of Mines, *S. Lany*, *J. Perkins*, *D. Ginley*, National Renewable Energy Laboratory

The rate of progress in the field of solar cells has been historically limited by the need for materials with desired functionality. Two complementary high-throughput approaches that have potential to facilitate such innovation are combinatorial thin-film experiments and predictive first-principles theory. Here we present examples of accelerated optimization of solar cell materials using the combined theoretical/experimental approach. The specific examples include (i) photovoltaic absorbers, and (ii) p-type contacts for solar cells

(i) PV absorbers are the key elements in any solar cells. Functionally, the absorbers are required to (1) absorb sunlight, and (2) facilitate extraction of charge carriers. We demonstrate accelerated progress towards (a) enhancement of optical absorption in Cu_2O, (b) improvement of electrical charge transport properties of Cu_3N, and (c) optimization of Cu-Sn-S material with respect to both optical and electrical properties. Our progress towards integration of these materials into thin film solar cell prototypes also will be discussed.

(ii) p-type contacts are needed for next-generation thin-film photovotlaics. Functionally such p-type contacts are required to (1) transmit sunlight, and (2) conduct holes. To accelerate the progress, we show (a) formulation of design principles (d6 oxide spinels) to guide the candidate selection [1, 2, 3], (b) down-selection of the most promising materials (Co_2ZnO4 and Co_2NiO4) from ~30 candidates using predictive theory [4], (c) optimization of the selected best-of-class materials (Co-Zn-O, Co-Ni-O) using thin-film combinatorial experiments [5], and (d) integration of the optimized materials (Zn-Ni-Co-O) as hole transport layers in organic photovotlaic devices [6].

In summary, combination of high-throughput theoretical and experimental methods demonstrated here can significantly accelerate the development of materials for thin film solar cells. This approach should be also suitable for discovery and optimization of materials for other technological applications.

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[1] V. Stevanovic et al Phys. Rev. Lett. 105, 075501 (2010)

[2] V. Stevanovic et al J. Am. Chem Soc. 133, 11649 (2011)

[3] J. Perkins, A. Zakutayev et al Phys. Rev. B 84, 205207 (2011)

[4] T. Paudel, A. Zakutayev et al Adv. Func. Mat. 21, 4493 (2011)

[5] A. Zakutayev et al Phys. Rev. B 85, 085204 (2012)

[6] A. Zakutayev et al MRS Comm. 1, 23 (2011)

10:40am **MG+EM+MI+MS-WeM9 The "Materials Genome" Project: Accelerated and Large-Scale Materials Discovery through Computation**, ***G. Ceder***, MIT **INVITED**

Novel materials design has become a critical capability to address several urgent societal problems. The need for novel materials is the technological Achilles Heel of our strategy to address the energy and climate problem facing the world, and over-reliance on critical elements has inspired large efforts to develop accelerated materials design strategies. The Materials Genome Project, originally started at MIT (www.materialsproject.com), has as its objective to use high-throughput first principles computations on an unparalleled scale to provide basic materials property data on all known and many potential new inorganic compounds, thereby facilitating the search for new materials.

I will show successful examples of high-throughput calculations in the field of lithium batterier, and show several new materials that have been discovered. I will discuss the public release version of the Materials Genome project which is making large quantities of computed data freely available to the materials community. The Materials Project is rapidly growing as a large collaborative environment for computed materials data.

11:20am **MG+EM+MI+MS-WeM11 Multifunctional Interfacial Materials by Design**, ***C. Eom***, University of Wisconsin-Madison **INVITED**

Complex oxides materials have been fertile ground for new discoveries, due particularly to their wide-ranging electronic, optical, and magnetic properties. Interfaces between complex oxides and related materials create juxtapositions between different symmetries and ordered states, and it has become clear that these interfaces are new materials in their own right and lead to dramatically different properties from those in bulk. But interfacial materials encompass a virtually unexplored territory, one in which theory or

experiment alone cannot be successful. New approaches must be implemented to understand basic principles, categorize competing interactions, and design and synthesize complex oxide interfaces with advanced functionalities. Our project focuses on an iterative cooperation between forefront theory and experiment that determines the fundamental principles controlling new physical phenomena at oxide interfaces, uses these principles to design couplings between multiple orders at interfaces to generate new functionalities, and experimentally synthesizes and investigates designed interfacial materials for novel electronic devices. These atomic-scale interfacial materials lead to, for example, new classes of electric-field controllable electronic and magnetic phenomena, and enable the development of new technologically important devices that exploit these couplings. Using a predictive theory and modeling, and feedback to theory from experiments, we have designed and synthesized novel oxide hetero-interfaces that have unique properties not presently available. This work has been done in collaboration with M.S. Rzchowski, C.J. Fennie, E.Y. Tsymbal, L.Q. Chen, X.Q. Pan, S. Ryu, T. Hernandez, T. R. Paudel, H. Zhou and D. D. Fong.

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