

ICAM-I2CAM & BOSTON UNIVERSITY PRESENT
DIGITAL DESIGN OF MATERIALS:
the way forward for materials science?

September 27–29, 2013
Workshop Abstracts

Friday, September 27

Introductory Overview

Gabriel Kotliar, Rutgers University

Session I: Solid State Chemistry

Discussion leader: David Coker, Boston University

- Evgeny Antipov, Lomonosov Moscow State University
How to discover new materials for energy application?

Ninety percent of the energy used today comes from fossil fuels, and this has serious consequences: rapid consumption of renewable energy sources, ecological damage, and climate change. There is therefore a need to increase the efficiency of use of fossil fuels and to develop clean, sustainable energy technologies.

Two classes of materials, which are widely studied for these technologies, will be presented and the current problems will be discussed.

1. Li-ion batteries originally developed for portable devices can now be found in applications as diverse as power tools, electric vehicles and stationary energy storage. To satisfy the need of current and new applications, Li-ion batteries require further improvement in terms of performance properties (energy and power density, safety and cost). Increasing the energy density that can be achieved in practical electrode materials would have a huge impact on all of the applications. This can be done through discovery and design of new materials for Li-ion batteries, or even more revolutionary, multivalent-ion batteries.
2. The electrode performance in the Solid Oxide Fuel Cells (SOFC) operating at elevated temperatures and exhibiting high efficiency depend on the electrode mixed ionic-electronic conductivity, its microstructure, its catalytic activity relative to the oxygen reduction, and the electronic band structures and chemistries of its bulk and surface regions. Understanding the fundamental, atomic-scale mechanisms for the oxygen electrocatalysis enhancement, will enable the systematic design and synthesis of novel nanostructured electrodes that exhibit the key properties which are important for practical application.

- Hideo Hosono, Tokyo Institute of Technology
Materials Design of Transparent Oxide Semiconductors

Materials frontier of transparent oxide conductors and semiconductors are expanding. So far materials were restricted to limited oxides, In_2O_3 , SnO_2 , ZnO , Ga_2O_3 , CdO and their complex compounds. I show several new materials which appears to differ rather from these materials including p-type materials and consider orientation of P/N carrier doping on the basis of band line-up we obtained.

- Ni Ni, University of California, Los Angeles
Progress in the study of high T_c electron doped $\text{Ca}_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ and $\text{Ca}_{10}(\text{Pt}_4\text{As}_8)(\text{Fe}_2\text{As}_2)_5$ superconductors

- Hidenori Takagi, University of Toky/MPI Stuttgart
Phase change functions in correlated transition metal oxides

I will briefly overview the concept of phase change functions of correlated electrons.

It is quite useful not only for electronics but also for a variety of applications.

Two case studies then will be given with emphasis on the strategy for improving the performance.

Using the relatively large entropy of correlated electrons, reflecting the spin/orbital degrees of freedom, electronic ice pack and thermoelectrics were developed.

The coupling of electronic phase change with lattice lead a novel strain functions such as giant negative thermal expansion.

The possible role of theoretical approach in designing materials and functions will be briefly mentioned.

Session II: The Search for Materials

Discussion leader: Malcolm Beasley, Stanford University

- Meigan Aronson, Stony Brook University
Electronic Delocalization in the Hunds Insulator LaMnPO: Implementing Theory Assisted Synthesis

- Zachary Fisk, University of California, Irvine
Where are the superconductors?

The heavy Fermions, cuprates, pnictides and even A15s reveal features common to the occurrence of their superconductivity. This question here is how can we use this to find new superconducting systems.

- Pascoal Pagliuso, Campinas State University (UNICAMP)
Routes for finding superconductors based on structural trends

We discuss experimental trends found for heavy fermions (namely the CeMIn₅ (M = Co, Rh, Ir) and related compounds) and FeAs-based superconductors that we related to key structural tuning parameters.

This allows us to establish simple criteria to search for new superconductors using crystal structures as a starting point. Digital design certainly is very valuable if it can reasonably predict if key physical properties are likely to be presented in a imaginary compound with selected crystal structures and chemical elements.

- Darrell Schlom, Cornell University
Exploring the Limits of Epitaxy to further Materials by Design

In this talk examples of the controlled epitaxial growth of oxides with perovskite, pyrochlore, rocksalt, Ruddlesden-Popper, and Aurivillius structures, including superlattices and metastable phases, are shown. Using epitaxy and the misfit strain imposed by an underlying substrate, it is possible to strain these complex oxides to percent levels—far beyond where they would crack in bulk. Under such strains, the band structure can be dramatically altered. Another way of altering the electronic structure is through dimensional confinement, either in ultrathin films or in natural (e.g., Ruddlesden-Popper) or artificial superlattices. The films are made by reactive molecular-beam epitaxy (MBE) and the emphasis of the talk will be on exploring the limits of epitaxy. For a few examples, the electronic structure measured on the pristine surface of the as-grown films by angle-resolved photoemission spectroscopy (ARPES), will be shown.

This work was done in collaboration with the groups of Kyle Shen (Physics, Cornell), David A. Muller (Applied and Engineering Physics, Cornell), and Reinhard Uecker (Leibniz Institute for Crystal Growth).

Saturday, September 28

Introductory Overview

Brian Sales, Oak Ridge National Laboratory

An Experimentalist's View of the Role of Theory in Moving Up the Materials Pyramid

The average time to develop a new material for a particular application is 18 years. A major driving force for the material genome initiative is to see if theory can be used to greatly shorten this time. The discovery/design of a new inorganic material can be pictorially described in terms of a "Materials Pyramid", where at the base of the pyramid are the many thousands of possible materials with known compositions and crystal structure types as typically reported by solid-state chemists. This extensive database forms the starting point for thinking about the discovery/design of new material with specific physical, electronic or magnetic properties. Very few materials make it all the way to the top of the pyramid where the material is used in a commercial product, but the economic impact can be enormous. In this presentation I will present an experimental point of view about how theory can accelerate this process, using examples from our group's recent research to illustrate some of the problems that occur in attempting to design functional materials without experimental input. What is most useful to experimental synthesis efforts are general phenomenological trends that provide simple guidelines or rules for how to think about large classes of materials. In my experience, theory is, at best, a compass that provides a rough direction for experimental synthesis, rather than a GPS-type result that points to a specific material. My personal opinion is that theory will only be useful in shortening the discovery time for a new material if it is closely coupled to an experimental program in an iterative fashion. Our group is currently attempting to use just such an approach to discover a new rare-earth poor permanent magnet.

Session III: State of the Art of Theory

Discussion leader: Karen Hallberg, Centro Atómico Bariloche

- Ryotaro Arita, University of Tokyo
Development of density functional theory for unconventional superconductors

Prediction or materials design of high temperature superconductivity is one of the holy grails of condensed matter theory. To achieve this goal, we need to develop a predictive method to calculate transition temperatures of superconductors (T_c). While density functional theory for conventional phonon-mediated superconductors was successfully formulated by E.K.U. Gross and others in 2005, it does not work for unconventional superconductors that often have extremely high T_c . As a first step toward the density functional theory for unconventional superconductors, we recently formulated a scheme for the plasmon mechanism, a prototypical unconventional pairing mechanism whose history dates back to late 1970s. We applied the new method to one of the most elemental high T_c superconductors, lithium under pressure, and obtained excellent agreement between theory and experimental data.

- Kristjan Haule, Rutgers University
Spin Dynamics and Pairing in Iron Superconductors from First Principles.

In a wide variety of materials, such as layered copper oxides, heavy fermions, organic salts, and iron pnictide compounds, superconductivity is found in close proximity to a magnetically ordered state. The character of this proximate magnetic phase is crucial for understanding the differences between the various families of unconventional superconductors and the mechanism of superconductivity. Using ab-initio approach Local Density Approximation + Dynamical Mean Field Theory (LDA+DMFT), we obtained charge dynamics (such as optical conductivity, and the anisotropy of the electronic states) as well as spin dynamics (dynamical structure factor and magnetic moments) in good agreement with experiment. Using

the ab-initio determined two-particle vertex function, we computed the pairing susceptibility in many families of iron superconductors, and we determined the most likely pairing gap symmetry in these compounds.

- Andrew Millis, Columbia University

Double counting, d-occupancy and the metal-insulator transition: the missing piece of a predictive theory of correlated electron physics in transition metal oxides

A full solution to the correlated electron problem is not feasible for materials or molecules of interest. Modern approaches apply advanced many-body methods to only a subset of electronic degrees of freedom while treating most of the electronic degrees of freedom via a computationally inexpensive mean-field-like method. A key issue in this approach is the self-consistent embedding of the correlation problem into the broader electronic structure. With C. Marianetti, H. Dang, H. Park, X. Wang and others I have investigated this issue in the context of the density functional plus dynamical mean field (DFT+DMFT) approach, where a key aspect of the embedding is encoded in the so-called "double counting correction". Here I will show that the standard DFT+DMFT approach (with full charge self consistency and the FLL double counting) wrongly predicts that several known Mott insulators are metals. This raises the question: are we solving the wrong many-body problem (by including in the correlation problem only d orbitals with onsite interactions), are we solving the right many-body problem wrongly (using single-site not cluster DMFT), or are we doing the embedding incorrectly. I will present results indicating that the dominant source of uncertainty is the double counting correction and suggesting ways it might be resolved.

- Mark van Schilfhaarde, King's College London

Predictive Nature of Self-consistent Quasiparticle Theory

- Cedric Weber, King's College London

An implementation of dynamical mean field theory for nano-structures and molecules

Phenomena that are connected to quantum mechanics, such as magnetism, transport, and the effect of impurity atoms and disorder, and their relation to material design and energy needs are important for almost every branch of the industry.

Density functional theory (DFT) was successful at making accurate predictions for many materials, in particular compounds which have a metallic behaviour. DFT combines high accuracy and moderate computational cost, but the computational effort of performing calculations with conventional DFT approaches is still non negligible and scales with the cube of the number of atoms.

A recent optimised implementation of DFT was however shown to scale linearly with the number of atoms (ONETEP), and opened the route to large scale DFT calculations for molecules and nano-structures.

Nonetheless, one bottleneck of DFT and ONETEP, is that it fails at describing well some of the compounds where strong correlations are present, in particular because the computational scheme has to capture both the band-like character of the uncorrelated part of the compound and the Mott-like features emerging from the local strongly correlated centres. A recent progress has been made in this direction by the dynamical mean-field theory (DMFT), that allows to describe the two limits (metal and insulator) in a remarkable precise way when combined with DFT.

The ONETEP+DMFT implementation and strategies to overcome the main bottlenecks of this type of calculations will be discussed, and its applications illustrated by a few case of studies.

References:

PRL 110, 106402 '13

Session IV: Tailoring Materials Using Mottness & Hundness

Discussion leader: Gabriel Kotliar, Rutgers University

- Dmitri Basov, University of California, San Diego

- Antoine Georges, College de France and École Polytechnique, France
Understanding and Controlling the Electronic Properties of Materials with Strong Correlations

Intra-atomic physics is key to the physics of materials with strong electronic correlations. Surprisingly, this is true even for rather itinerant metals, such as the transition-metal oxides of the 4d series, e.g. ruthenates. These materials are not close to a Mott insulating state, but nevertheless display strong correlations because of the intra-atomic Hund's coupling.

The self-consistent embedded-atom construction at the heart of Dynamical Mean-Field Theory proves to be essential for understanding these materials.

Reference: A.Georges, L. de'Medici and J. Mravlje.
Annual Reviews of Condensed Matter Physics, Vol.4 (2013) arXiv:1207.3033

- Marcelo Rozenberg, CNRS
Universal electric-field-driven resistive transition in Mott insulators and A digitally inspired prediction of novel emergent behavior in correlated materials

- Yasutomo Uemura, Columbia University
Novel DMS ferromagnets $\text{Li}(\text{Zn},\text{Mn})\text{As}$, $(\text{Ba},\text{K})(\text{Zn},\text{Mn})_2\text{As}_2$ and $(\text{La},\text{Ba})(\text{Zn},\text{Mn})\text{AsO}$ isostructural to FeAs superconductors

We have synthesized novel diluted ferromagnetic semiconductor (DMS) systems $\text{Li}(\text{Zn},\text{Mn})\text{As}$ (T_c up to 50 K) [1], $(\text{Ba},\text{K})(\text{Zn},\text{Mn})_2\text{As}_2$ (T_c up to 200 K) [2] and $(\text{La},\text{Ba})(\text{Zn},\text{Mn})\text{AsO}$ (T_c up to 30 K) [3], which have similar or identical crystal structures with the 111, 122 and 1111 FeAs superconductors, following a theoretical proposal of Jungwirth and collaborators [4] on $\text{Li}(\text{Zn},\text{Mn})\text{As}$. Unlike $(\text{Ga},\text{Mn})\text{As}$ which is available only as MBE-produced films due to limited chemical solubility for $\text{Ga}^{3+}:\text{Mn}^{2+}$ substitutions, the above mentioned systems were obtained in bulk form via iso-valent $\text{Zn}^{2+}:\text{Mn}^{2+}$ substitutions. Spin doping is independent from charge doping in these novel DMS systems unlike $(\text{Ga},\text{Mn})\text{As}$ available only as p-type systems due to simultaneous spin and charge dopings. Encouraged by a very good matching of lattice constants and identical crystal structures of semiconducting BaZn_2As_2 , ferromagnetic $(\text{Ba},\text{K})(\text{Zn},\text{Mn})_2\text{As}_2$, antiferromagnetic BaMn_2As_2 and superconducting $(\text{Ba},\text{K})\text{Fe}_2\text{As}_2$, we hope to develop junction/multi-layer spintronics devices consisting of various combinations of these ground states. Most of the FeAs superconductors may have their companion DMS ferromagnets, made not only of As compounds but also of P, Sb, and others with various ways of charge dopings. For materials development in this situation, computational approach based on LDA, DMFT, Quantum Monte Carlo and other methods will be very helpful.

- [1] Z. Deng et al., Nature Communications 2 (2011) 422
- [2] K. Zhao et al., Nature Communications 4 (2013) 1442
- [3] C. Ding et al., Phys. Rev. B88 (2013) 041102(R)
- [4] J. Masek, J. Kudrnovsky, F. Maca, B.L. Gallagher, R.P. Campion, D.H. Gregory, and T. Jungwirth, Phys. Rev. Lett. 98, 067202 (2007).

Work performed in collaboration with the groups of
 C.Q. Jin (IOP Beijing), F.L. Ning (Zhejiang), G.M. Luke (McMaster),
 P. Boeni (TU Munich), E. Morenzoni (PSI), S. Maekawa (JAEA)
 with financial supports by NSF-PIRE (Partnership
 for International Research and Education) OISE-0968226 and
 DMR-1105961, and JAEA Reimei Project.

Sunday, September 29

Session V: Novel Materials

Discussion leader: David Campbell, Boston University

- Massimo Capone, CNR-IOM and SISSA
When Mott meets BCS: Molecular Conductors and the search for high-T_c superconductivity

There is a widespread tendency to divide superconductors in two broad families: Standard superconductors in which superconductivity is due to electron-phonon interaction and exotic superconductors where pairing arises from purely electronic interactions. In the second family, obviously dominated by the high-T_c cuprates, the proximity to a Mott insulator is believed to be the key element underlying the phase diagram.

Molecular superconductors like the fullerides challenge this distinction. The molecular nature leads indeed to narrow bands which imply strong correlation effects, while the coupling to molecular phonons is strong and can give rise to pairing. We show that the two effects can be cooperative, leading to a phononic superconductor which is actually favoured by strong correlations and Mott physics [1]. Despite the phononic nature, the systems strongly deviate from the Bardeen-Cooper-Schrieffer predictions in many ways. Cs₃C₆₀ is indeed the realization of this scenario [2]. The material is a Mott insulator at ambient pressure, but it turns into a superconductor with T_c up to 38K under pressure. The superconducting T_c has a bell-shaped behavior as a function of pressure that reminds of that of cuprates as a function of doping.

We finally discuss the relevance of these results for other molecular (super)conductors, and the generality of the paradigm we have identified.

[1] M. Capone, M. Fabrizio, C. Castellani and E. Tosatti, Rev. Mod. Phys. 81, 943 (2009); Science 296, 2364 (2002)

[2] Y. Takabayashi et al., Science 323, 1585 (2009); A.Y. Ganin et al. Nature Materials 7 367 (2008)

- Kenneth Burch, Boston College
New Properties by Treating Materials like Legos

- Matthew Rosseinsky, University of Liverpool
Identification of New Functional Materials – Integration of Experiment and Computation?

The development of advanced materials will increasingly rely on our ability to assemble complex compositions in an ordered and predictable manner. The interplay of computation and experiment offers insight into the structures and compositions accessible in both thin film (1) and bulk (2) materials. I will present an example of crystal chemically-informed computational identification of a new SOFC cathode

(3), and an example of a nanostructured SOFC cathode system (4) whose multiple length scale structure exemplifies the challenges in designing materials ab initio.

- (1) M. Dyer et al, *Angewandte Chemie International Edition* 51, 3418, 2012
- (2) J. Rabone et al *Science* 329, 1053, 2010
- (3) M. Dyer et al *Science* 340, 847, 2013
- (4) A. Demont et al *J. Am. Chem. Soc.* 135, 10114, 2013