Breakout Report on Correlated Materials

Identification of Grand Challenges

Introduction

In celebration of the 2-year anniversary of the Materials Genome Initiative (MGI), referred to as '*Predictive Theory and Modeling for Materials and Chemical Sciences*' by BES, a workshop was held to begin the process of identifying 'grand challenges' for which an integrated theory-computational-experimental approach would greatly accelerate progress in the field. The MGI is a cross-government initiative that includes DOE, NSF, DOD, and NIST.

Materials Genome Initiative

The Materials Genome Initiative is a new, multi-stakeholder effort to develop an infrastructure to accelerate advanced materials discovery and deployment in the United States. Over the last several decades there has been significant Federal investment in new experimental processes and techniques for designing advanced materials. This new focused initiative will better leverage existing Federal investments through the use of computational capabilities, data management, and an integrated approach to materials science and engineering.

http://www.whitehouse.gov/mgi

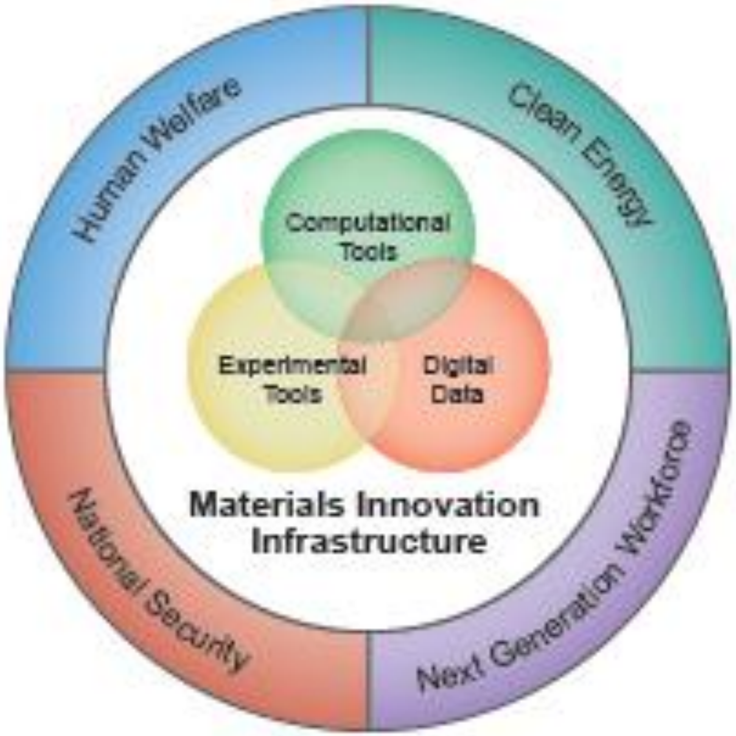


Figure 3: Initiative overview

Five topic areas

Light Weight & Structural Materials

Electronic & Photonic Materials

Energy Storage

Catalysts

Correlated Electron Materials

Meeting Timeline

- 6/25 Breakout plenaries
- 6/25 Brainstorming on Grand Challenges, keeping in mind the following three questions
  - *“If materials scientists could ________, then new pathways of ________ materials discovery would be possible.”*
  - *“If materials scientists could ________, materials/product engineers would be able to ________.”*
  - *"Materials/product engineers need to be able to ________, which materials scientists could enable by _________."*
- 6/26 AM Brainstorming—Sort ideas into categories, refine
- 6/26 AM Brainstorming—Priorities
- Load into this PPT

Breakout - correlated materials

- Chairs: Littlewood (Argonne), Parkin (IBM)
- Speakers: Terris (HGST), Kotliar(Rutgers)
- Scribe: Lau (NIST)
- Participants:

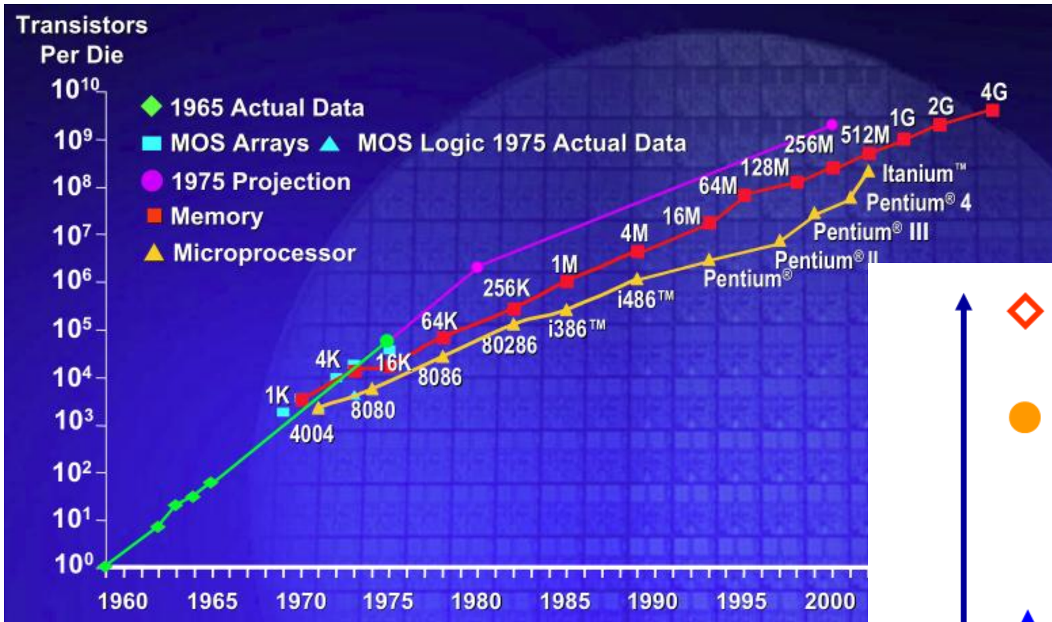
  Bansil (Northeastern), Childress (HGST), Devereaux (SLAC), Hill (NHMFL), Johnson (Ames), Kryder (Carnegie Mellon), Larbalestier (FSU), Malozemoff (American Superconductor), Morgan (Wisconsin), Mryasov (Alabama), Norman(ANL), Sefat (ORNL), Selvamanickam (Houston), Shelton (PNNL)

Correlated materials

- Not just emergent properties (magnetism, superconductivity)
- Battery electrodes (ideal Li-ion cathode is a Mott insulator)
- (Electro-) chemistry at interfaces
- Combustion chemistry

Engineering driven modeling and characterization tools

The consequence of understanding is prediction: Moore's Law for Si vs. current strategy for Li-ion batteries



◇ **Unk-HV-HC / Li metal**

Safe and reversible cycling of Li metal
Market entry >2021

● **Unk-HV-HC / Gr-Si**

Discovery of high voltage electrolyte >4.8 V
Discovery of reversible unknown high-voltage high-capacity cathode: 250 mAh/g @ 4.8 V
Market entry > 2019

△ **Li_2MXO_4 / Gr-Si**

Discovery of path to reversible multi-electron cathode material with 4V cell voltage
Market entry > 2017

■ **LMR-NMC / Gr-Si**

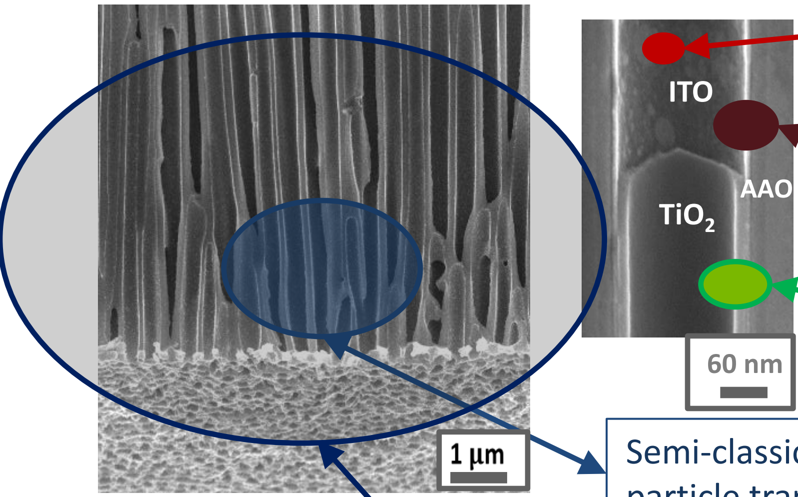
Stabilization of silicon
Market entry > 2015

◇ **LMR-NMC / Gr**

Stabilization of LMR-NMC
Market entry > 2013

● **LMO / Gr**

Computational Chemistry and Materials Science: designing what you make



Prediction of new materials and structures on the atomic scale, including interfaces, growth and defects

Excited state calculations for electron transfer and photon-mediated transitions

Accurate intermolecular potentials to model structure and dynamics on nanoscale

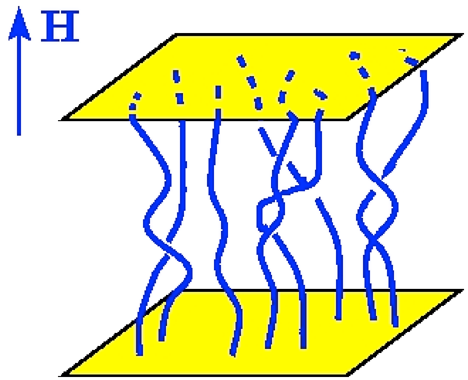
Semi-classical models of electrical and particle transport on mesoscale

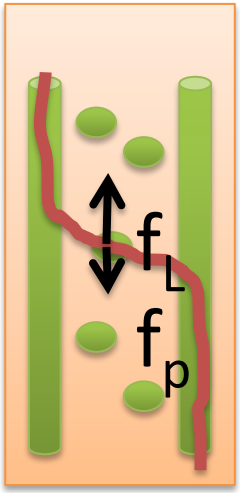
Effective theories of inhomogeneous media: elastic, fluid and electrical transport

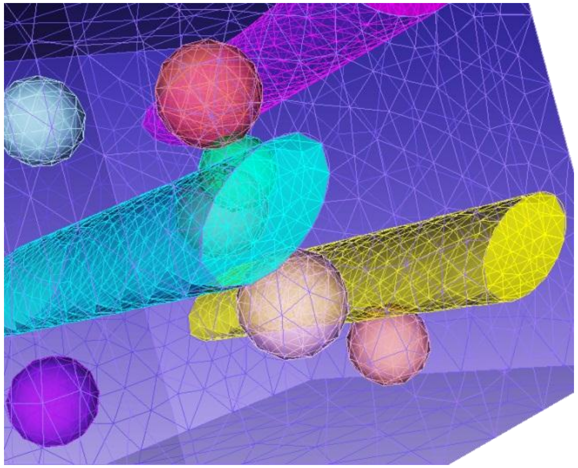
- Each box requires new investment in methods, theory and computation
- Joining up the boxes is as important as the investment in any single piece
- We must curate both data and software
- Design choices driven by application target

Demands a collective corporate effort linking computation, methods, software, and data guided by an engineering goal

Better superconductors - design of vortex pinning for large current applications







$$\mathcal{F}_{\mathrm{GL}} = \frac{1}{2}\int d^d x \left\{ \beta\left(\frac{\alpha}{\beta} + |\psi|^2\right)^2 + \frac{\hbar^2}{m}\left|\left(i\nabla - \frac{2\pi}{\phi_0}\mathbf{A}\right)\psi\right|^2 + \frac{1}{4\pi}(\nabla\times\mathbf{A} - \mathbf{H})^2 \right\}$$

Time-dependent Ginzburg -Landau eqn.

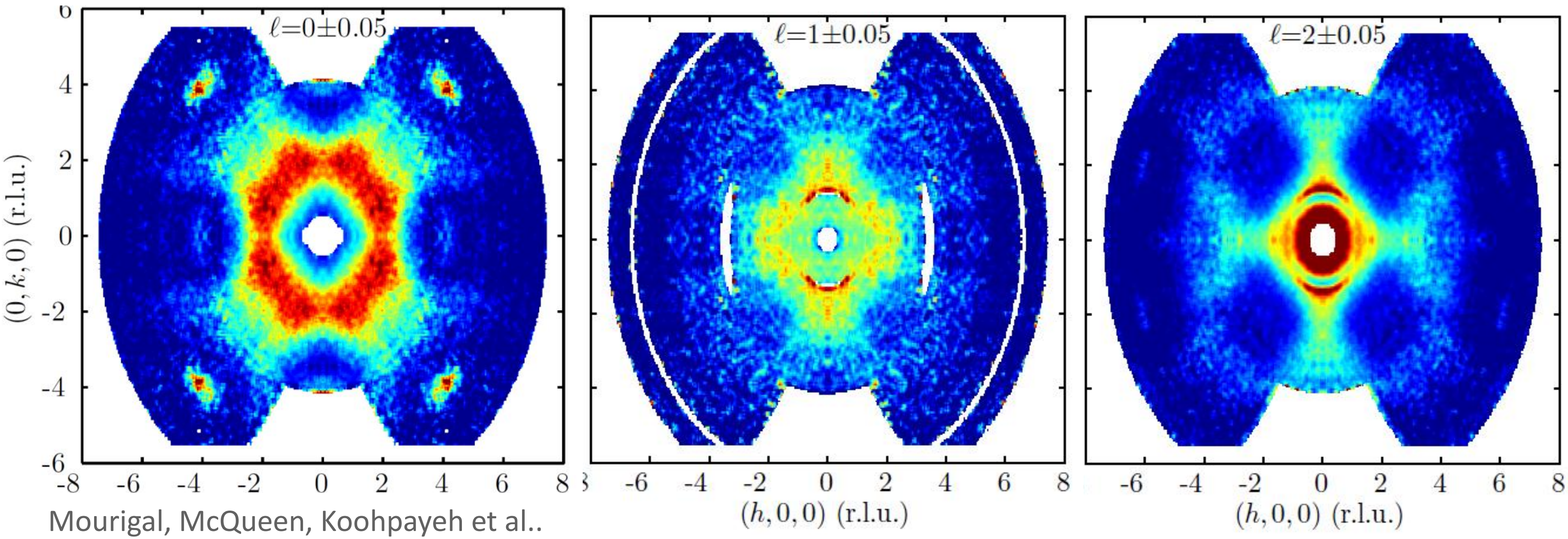
$$\frac{\partial\Psi}{\partial t} = -\frac{\delta\mathcal{F}_{\mathrm{GL}}}{\delta\Psi^*}, \quad \frac{\delta\mathcal{F}_{\mathrm{GL}}}{\delta\mathbf{A}} = 0$$

Equations well understood: but contain long range forces, disjoint length scales, and need long times

BES-SCIDAC – Andreas Glatz, Argonne

Theory and experiment meet around "big data"

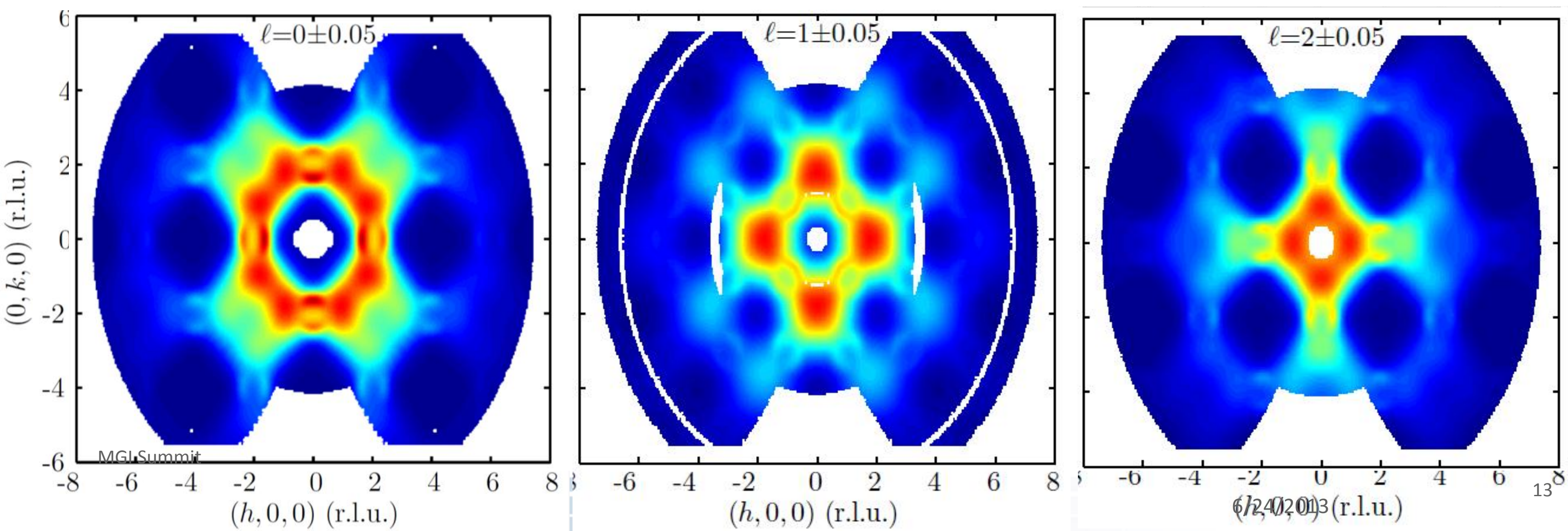
Spin fluctuations in a quantum paramagnet (Collin Broholm)



Mourigal, McQueen, Koohpayeh et al..

Large N expansion: Nearest Neighbor Interactions only (Chalker and Conlon)

$J_2=0.2J_1$, $J_{3a}=0.2J_1$, $J_{3b}=-0.01J_1$ (Chalker and Conlon)



Building multiscale models via “genomics”

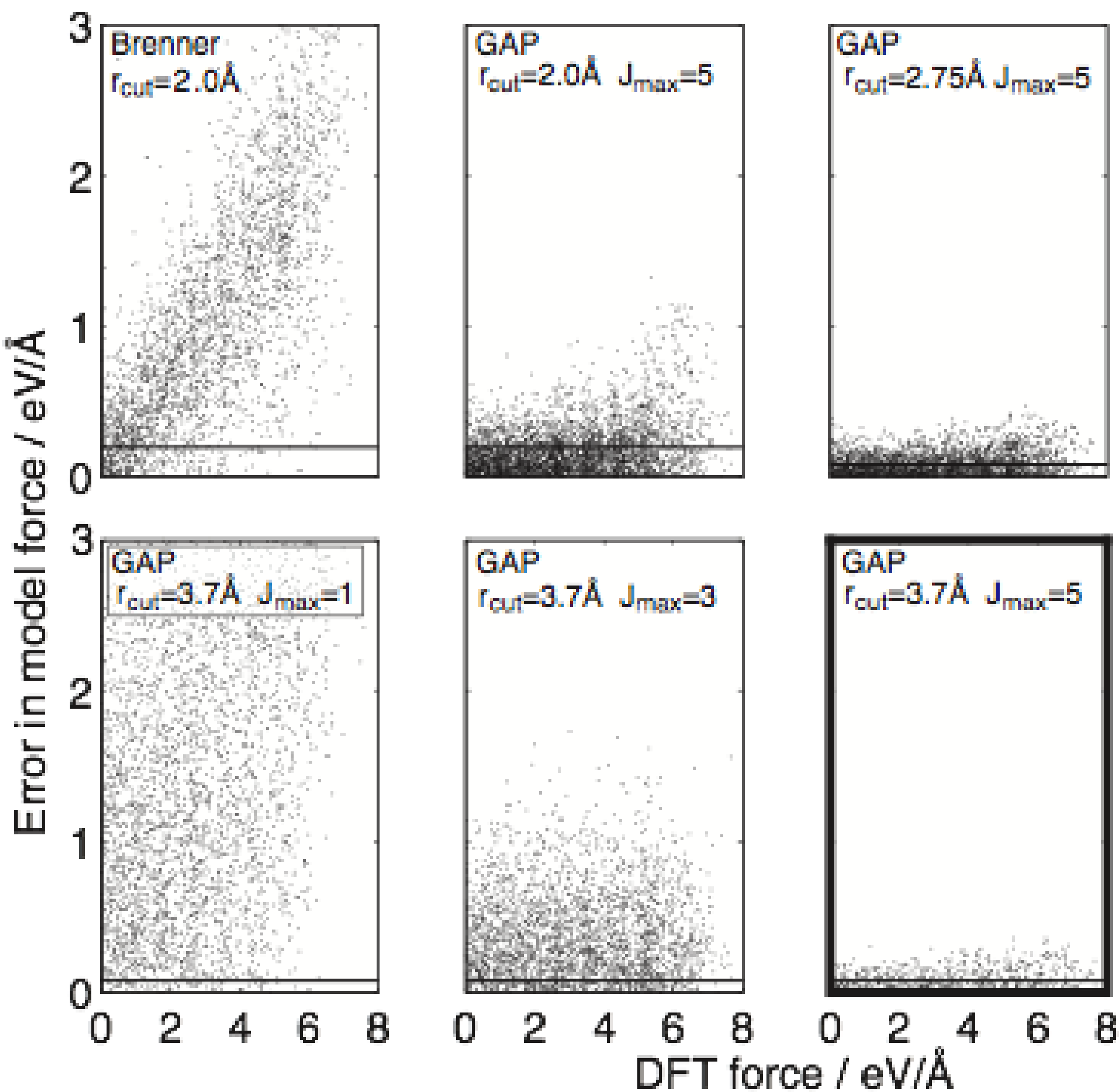
Materials by design: genomics?

Genomics must be grounded in theory: the human genome initiative depends fundamentally on the “central dogma” of DNA coding. This is both the fundamental **theory** of biology and an **algorithm**

Materials genomics derives its validity from the Schrodinger equation – but this is not (yet) an instruction set

Robert Laughlin (Nobel lecture)

Supervised learning: Gaussian Approximation Potentials trained on DFT (Gabor Csanyi)



Diamond
(1000 K)

Theoretical limit
for given cutoff:
RMS of
long range forces

Energy error < 1 meV (0.02 kcal/mol) / atom

Grand Challenges

Modelling materials structure, growth, patterning

A common refrain was that it is often possible to calculate functional properties with appropriate accuracy if the position of the atoms is known. Often properties depend critically on interfaces, surfaces, grain boundaries, and defects, which are often not well-understood, and not well-modeled.

- **Impacts**: Practical examples include textured substrates for magnetic deposition, textured superconducting wires, interfaces between magnetic thin films, surface structure of catalysts, solid-electrolyte interface. A corollary is that using modeling to aid synthesis and growth, as well as developing models for materials processing at the nanoscale (e.g. etching) was thought to be a way to speed up the manufacturing design process.
- **Requirements**: scale-bridging methods; in situ measurements of structure during growth; close theory/experiment partnership

Rapid surveying of correlated materials

While ab initio methods for correlated materials are not at the level of DFT codes where MGI can attack the inverse problem, tools that are good enough to produce trends in formation energies, structure, and excitations are emerging, including methods such as hybrid functionals, GW, DMFT, QMC. Materials needs are often matched to multiple properties, so multi-variate optimization is necessary.

- **Impacts**: Guided synthesis of new materials classes; sifting through complex materials phase space using multi-variate optimisation; candidate materials classes for new thermoelectrics, magnets, electrodes, …
- **Requirements**: Methods development; verification and validation; curation of data;

Sub 10 nm device fabrication – toward a nano-3D printer.

While this is the most extreme goal, the current focus on 2D deposition with atomic accuracy is something that gradually needs to give way to full 3D structures.

- **Impacts.**
  - Higher density magnetic recording, memory, and logic than anything we have today.
  - Present-day lithography, a subtractive process, is expensive, labor intensive, requires huge capital investment, and wastes a lot of process material.
  - Nano 3D printing may(?) not be useful for end manufacturing, but it would be wonderful for research and rapid prototyping
- **Requirements.**
  - Non-He temperature manipulation of atoms
  - A theory of etching

Full system modeling of complex devices

System models would have to integrate from the nano-scale upward, bridging scales, and methodologies. While some applications are clearly further out than others, those that operate in a fairly tight design space (e.g. a combustion engine, a magnetic read head, a superconducting multilayer tape) could be the first targets

- **Impacts.** A virtual battery model, a virtual combustion engine, a virtual read-head, a virtual deposition system would all be extraordinary design tools that would accelerate product development and reduce cost.
- **Requirements.** Open source codes with workflow control ; tight integration of engineering/measurement/simulation; quantitative schemes for scale-bridging

Simultaneous/Integrated simulation and experiment

The current scientific model where theory and experiment run sequentially can be slow, and is one of the bottlenecks of the data deluge. In future it may not be possible to collect “all” the data for subsequent analysis, but even small experiments could benefit from integrated hypothesis-testing. Some of the earliest opportunities for impact could come at large facilities, where some experiments generate large 4D datasets

- **Impacts.** Speed up in the discovery process; more effective use of beam time
- **Requirements.** Focus on detector science/technology/data. Computation integrated with I/O

New devices by control of correlated phenomena

The on/off ratio of a conventional semiconductor device is controlled by the ratio of temperature to the energy gap, which is in principle an unnecessary – but in practice huge – constraint. Correlated materials offer in principle a much wider palette, but we have yet to gain *reliable* external (e.g. by electrical bias, strain, optical methods,…) control of metal insulator transitions, or more complex phenomena. Opportunities here include interface engineering in oxides, nano-scale control of electrochemistry, strain engineering of thin films, defect engineering for nonlinear memory devices.

- **Impacts.** Entirely new product lines in low power electronics, sensors and detectors, high power electronics (WBG replacements), new memory devices,
- **Requirements.** Focus on control and measurement at the nanoscale. Predictive modeling of correlated materials validated by in situ measurement tools.

Delivery Mechanisms

- First do no harm/moderation in all things – several felt that the individual PI culture was at the core of US creativity and should not be imperiled. So the goal of collective activities must be to *support* PI's.
- Mini hubs (Project focused or platform focused? Materials focused? Not instrument focused? Vertically integrated?). Seen as successful but the need for multiple styles was felt
- Materials design centers, focused on particular classes of materials. Consensus on need for expansion in fundamental materials exploration and discovery
- Network of science focused (NSF?)/materials focused (BES?)/technology focused (NIST) and vertically integrated (EERE/DOD) hubs.
- Software/method development centers – integrate BES/ASCR user facilities? Many of the core needs which were identified to support grand challenge activities require the support of open source methods, verification and validation, and data curation, in order to support a potentially very large user community that includes industry and experiment.
- Mechanism for open source software development/support
- Real-time theory/simulations and meta data/ data capture at user facilities
- Mechanism for data capture from existing and published work
- "Google materials" free-access "social media" for materials scientists

Collation of answers to specific questions

Engineering pull

Materials/product engineers need to be able to __A_______, which materials scientists could enable by ___B_______.

A

- Make textured materials
- Control materials growth
- Engineer vortex pinning in superconductors
- produce layered combination of materials with large resistive response , controllable anisotropy of magnetic layers
- Defect control in oxides
- Sub 10-nm device fabrication
- Nano 3D printer

B

- Multiscale modeling
- In operando theory
- Connect ab initio to GLAG theory
- spin dependent transport simulations, rational design of magnetic anisotropy
- appropriate models/theory of defects and interfaces
- Theory of etching
- Develop advanced ALD

Science push: Materials discovery

If we could do __A_, it would make possible _B___ new pathways of materials discovery.

A

- Develop “good enough/robust” models for structure, defects excitations, transport across scales
- Predictive capabilities for correlated (solid/solid or solid/liquid) interfaces
- Rapidly survey strongly correlated materials
- Make facile connection across length scales
- model sputter growth of "real" multiple layer materials
- Simultaneous experiment/theory feedback
- New materials by non-equilibrium processes.

B

- Thermoelectrics, battery electrode materials, magnets, superconductors, topological insulators ….
- Process control
- rapidly accelerate development of new thin films for a variety of technologies

Science push: Product development

If we could do _______A______, materials product engineers would be able to ______B___.

- Validated, open source codes with workflow control
- Materials-specific informed scale-bridging.
- Find a cubic superconductor with T_c > 100K, large J_c
- Design/control metal insulator transitions with external control
- Design surface binding of small molecules
- Multi-variate optimisation of materials systems parameters
- Design/predict/fabricate higher soft Bs magnets
- Higher Bs hard magnets
- Modeling finite temperature properties of real materials.
- Annealing behavior of materials

- Accelerated in house design. Will generate jobs.
- 77K magnets and lightweight motors, and magnet cables ,low cost.
- New sensor, logic and memory devices; better cathodes.
- Catalysts, biofuels, metal-air batteries, methane conversion
- Simplify materials choices, would reduce time and cost to product production.
- increase HDD density 10x with current head technology
- rare-earth magnet replacement