The Outstanding Challenge of 21st Century Science that can be Addressed Using Neutron Scattering

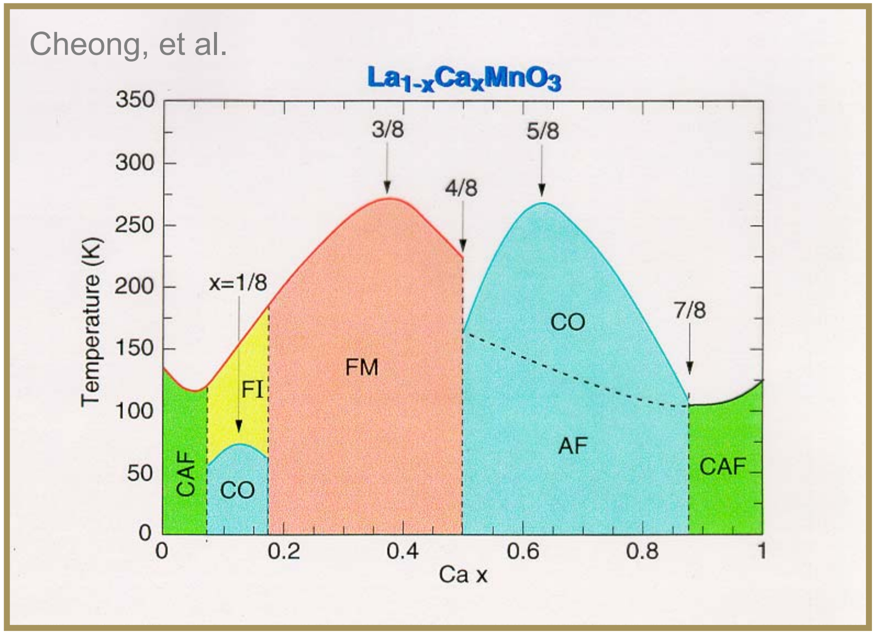
- Need to UNDERSTAND self-organizing behavior that first emerges on the nanoscale in chemically complex systems
- USE self-organization to create materials / systems with greatly enhanced or new combinations of properties

"CORRELATED ELECTRON" MATERIALS

- Discoveries of many important properties not understood using traditional ideas
  Copper-oxide high-temperature superconductors; metallic NiO; colossal magnetoresistance (CMR) in transition metal oxides; heavy-Fermion metals (rare earths and actinides); 1D and 2D electron gases; organic charge-transfer compounds

- CHARACTERISTIC: COEXISTENCE of several different kinds of ORDER
  -- Charge, orbital, spin density, super-conductivity, magnetism
  -- COMPETING or SYNERGISTIC ordering ?
  -- Mathematical description being developed
     Field theory: ground & lowest excited states
     Numerical calculations: leadership computers + new, efficient algorithms

- ORIGIN: Breakdown of the independent-electron approximation
  Strongly interacting electrons behavior is "highly correlated"



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Importance of the Challenge and of Neutron Scattering

"Clearly, highly correlated electron systems present us with profound new problems that almost certainly will represent deep and formidable challenges well into this new century…"

"…neutron scattering is an absolutely indispensable tool for studying the exotic magnetic and charge ordering exhibited by highly correlated electronic systems."

R. J. Birgeneau and M. A. Kastner, Editorial, *Science* 288, 437 (21 April 2000)

But What is Needed and Why?
What are the Current Limitations for Neutron Scattering?

SINGLE-CRYSTAL SAMPLES FOR THE MOST REVEALING EXPERIMENTS

- Reflectometry, Diffraction (structure), Inelastic (dynamics)
- Inelastic neutron scattering: Elementary excitations and dynamics
  -- Requires significant sample mass: 50-100 mg at SNS
- (Birgenau & Kastner) Japanese leadership in correlated electron materials research due to high value placed on single-crystal growers in Japanese universities

SERIOUS LIMITATIONS OF CONVENTIONAL “BULK” CRYSTAL GROWTH

- Chemical composition, temperature, pressure are the only variables
- Limited to “naturally” occurring equilibrium phases → *unable* to enhance, “tune”, or create the most interesting exotic properties
  -- Example: multiferroics, e.g., coexistence of ferromagnetism and ferroelectricity
  -- Few “natural” multiferroics and only weakly ferroelectric

Crystal growth methods are needed that enable NANOSCALE MODIFICATION and CONTROL of self-organization and ordering

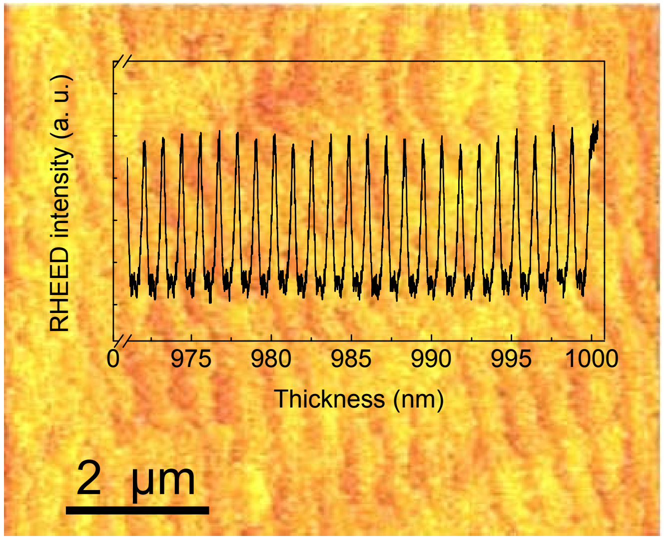
An Enabling Discovery: Growth of Thick "Superlattice Crystals" for Forefront Neutron- and Nano-Science

- **PLD: the method of choice for *oxide* films and heterostructures growth**

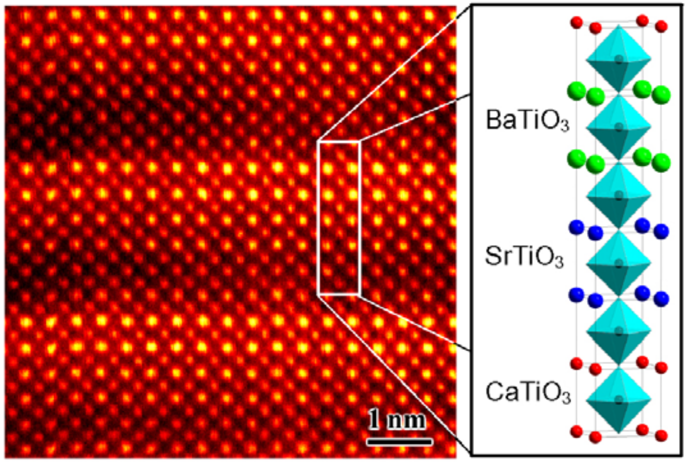
- ***New understanding + new methods* for pulsed laser deposition (PLD) now enable growth of THICK oxide heterostructures suitable for ALL neutron scattering methods**

- **Example: Growth of 1 μm-thick structure containing thousands of unit cells with**
  -- atomic-layer control
  -- complete reproducibility of surface and interfacial quality

- **DEMONSTRATION: *First experimental verification* of theoretically predicted polarization enhance-ments in *artificial PLD ferroelectric "superlattice crystals"* grown from $SrTiO_3$, $BaTiO_3$, and $CaTiO_3$ building blocks**
  [H.N. Lee et al., *Nature* 433, 395 (2005), and Materials Science News and Views, *Nature* 433, 369 (2005)]



RHEED oscillations used to monitor growth of unit cells #975 - 1000



Artificial ferroelectric-dielectric superlattice $[(BaTiO_3)_2/(SrTiO_3)_2/(CaTiO_3)_2]$, with compositionally-abrupt interfaces as seen using Z-contrast TEM.

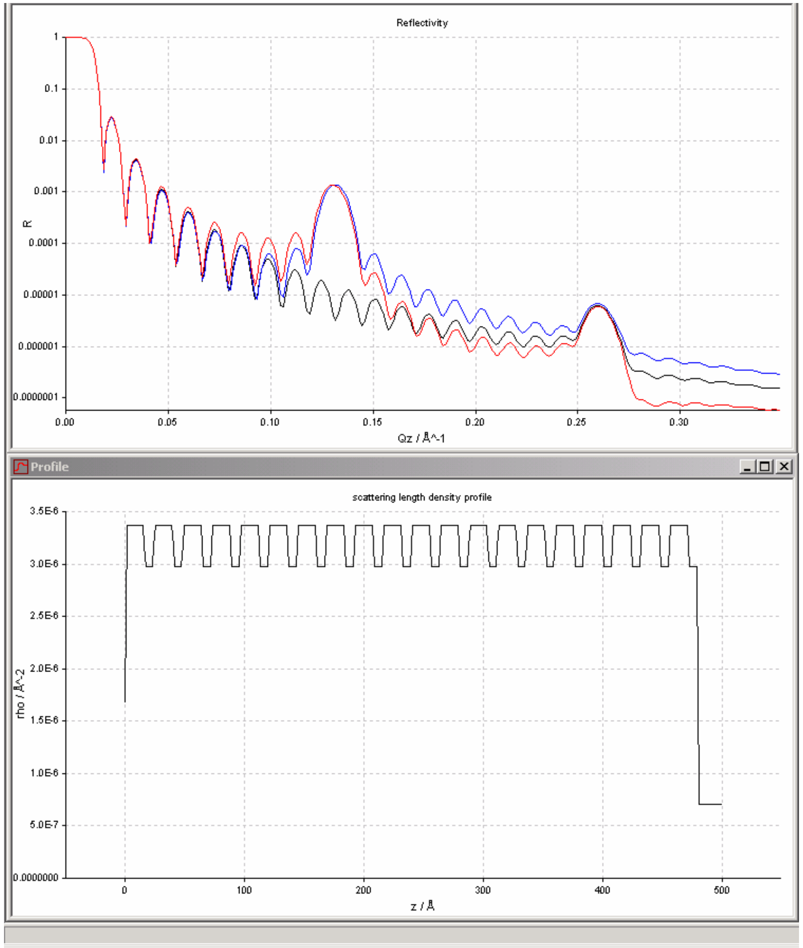
Simulations and Experience-based Estimates

Growth of Artificial "Superlattice Crystals" for the Full Range of Neutron-Scattering Experiments Now Feasible

THICK OXIDE HETEROSTRUCTURE GROWTH ESTIMATES

- **GOAL: Sample mass sufficient for high-resolution *inelastic* neutron scattering**
- **Area = 10 mm x 10 mm and 100 μm thick**
  - **→ volume = 10 mm^3 and mass ~ 50 mg**
  - **→ sufficient for detailed data analysis**
- **Sample growth duration < 4 days**
  - **-- 100 Hz laser, 100 pulses / unit cell**
  - **-- comparable to bulk crystal growth**
- ***Diffraction*: 20 mg sufficient for complete structure determination in < 2 hrs @ SNS**
- ***Reflectometry*: Only 0.5 – 5 μm thickness for S/N ~ 10-100 @ large Q, R_{min}~ 10^{-6} (typ.)**
  - **-- Example: determine magnetic polarization directions in adjacent superconducting & magnetic layers of superlattice**

Neutron reflectometry: 20-period superlattice, antiferromagnetic ordering



Outstanding Opportunity and Need to Build a
First-of-its-Kind "Superlattice Crystal" Growth Facility

- **Move neutron scattering beyond limits of conventional bulk crystal growth**
- **Novel properties result from *competition* between physical dimensions of nanostructures and length scales characteristic of collective phenomena**

  → capability for *nanoscale control* of dimensions of individual components in multilayered structures *is essential*

- **"*Design* of novel samples should be an integral part of the planning process for neutron scattering." (Paul Canfield, Ames Laboratory, NHMFL workshop)**
- ***Rich opportunity!* Lattice-compatible oxides include insulators, conductors, ferro- and antiferro-magnets, high-temperature superconductors, CMR compounds, and ferroelectrics**
- **What will be properties of an artificial "superlattice crystal" constructed from ferro*electric* and ferro*magnetic* building blocks?**
- **Future direction: strongly *multiferroic* oxide "designer crystals"**

BENEFITS

- **Fully engage international theoretical and experimental communities in *designing* materials to *create new phenomena***
- ***Greatly enrich the data stream* resulting from national investment at SNS**

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