2nd International Workshop on Software Solutions for Integrated Computational Materials Engineering
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Sandbox Scenario 3

Fabrication of single crystal thin film of ferroelectrics

Introduction

Electronics is ubiquitous in almost every technology device and nowadays can be found in communication systems (radios, transmitters, recorders etc.), data processing and storing systems (computers and measuring instruments) and control systems (in machines, airplanes, power stations). Ferroelectrics is an important device in today's world, as capacitors, for example in camera flashes, and as tunable microwave device, that exhibits spontaneous electric polarization that can be reversed by the application of an appropriate electric field. In thin film form, it is important for non-volatile memory storage. While significant advances have occurred in deposition of thin films of ferroelectrics, high-quality single-crystal thin-films are non-trivial to obtain because it requires lattice matching to the growth substrate, yet these films may still lack some properties of bulk ferroelectrics. A novel crystal ion slicing has been developed to obtain single-crystal thin-films, already demonstrating on those transition metal oxides including $SrTiO_3$.

Discovery and fabrication of single crystal thin film of ferroelectrics

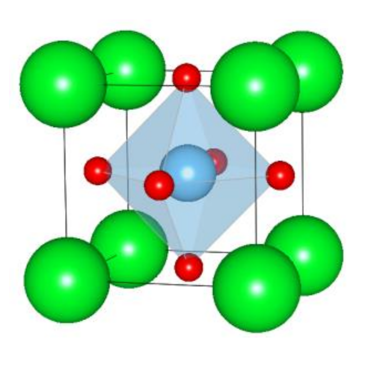

Figure 1: The crystal structure of perovskite ferroelectrics.

Ferroelectric materials have a perovskite (Figure 1) or related crystal structure, many of which show also good piezoelectric. By combining computational thermodynamics and electronic-structure methods (DFT, Molecular Dynamics, etc.) with intelligent data mining and database construction for searching novel transition metal oxides for desirable ferroelectric candidates.

High throughput (combinatorial) experimentation has emerged as a relatively new paradigm that offers rapid and efficient materials including ferroelectrics structure verification, screening and optimization. It is characterized by synthesis of "sample library", either continuous or discrete, that typically contains the materials variation of composition and state-of-the-art local micro-analytical measurement that results in massive datasets.

High quality perovskite (like strontium titanate) in single crystals form can be grown by a floating zone method with radiation heating. Additional crystal heating below the molten zone by an in-growth annealing furnace is designed to lower the temperature gradients in order to achieve slower cooling of the grown crystal. The perfect crystals can be grown reproducibly with starting materials of 4N grade quality.

Thin complex perovskite films (<1 µm thick) can be directly fabricated from a single-crystal bulk sample by using ion slicing. The process, based on ion implantation and anodic bonding, separates thin films having from bulk crystals. The dielectric properties and crystal structure of such thin films are found to be essentially those of the bulk single crystal.

In short, a non-trivial ICME methodology for discovery and fabrication of single crystal ferroelectric thin films, consisting of four process steps, is present for $(Ba,Sr)TiO_3$ compounds in Figure 2.

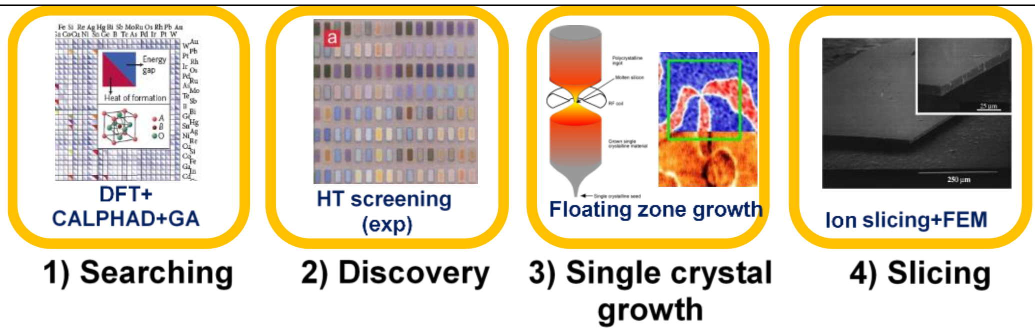

Figure 2: Discovery and fabrication of single crystal thin film of ferroelectrics.

The process chain is of complex multiphysics and multiscale, involving the spatial and temporal inhomogeneity in composition, variant, polarization, twinning, etc.

1. **High throughput computational quantum (DFT), thermodynamics (CALPHAD) & mining (GA) for searching multinary compounds:**

Can we identify good candidates of multinary ferroelectrics over the possible wide composition space (arrays) completely through high throughput computational design of, e.g.

- Crystal structure,

- Atomic structure imperfection,
- Phase stability,
- Key material properties (esp. dielectric) needed for guiding the subsequent process steps.

2. High throughput discovery and verification of oxides:

Can we experimentally perform rapid screening of such critical materials properties as

- Dielectric constant and tunability,
- Dielectric loss,
- Spontaneous polarization,
- Coercive field,
- Breakdown voltage.

3. Single crystal (bulk form) growth with its resultant microstructure & related mechanism:

The growth process is of complex multiphysics. Can we predict, e.g. by mesoscale modeling like phase field or Landau theory, the temporal and spatial inhomogeneity of single crystal ferroelectrics in:

- Transition metal dopants,
- Variant and twinning of product phase,
- Residual stress/strain,
- Materials properties (inc polarization, dielectric constant, and Breakdown voltage, etc.)
- Gradients in properties.

4. Ion slicing for ferroelectric components and meso/macro mechanics (FE like COMSO).

Can we predict the multiscale process of ion slicing?

- Dislocation density,
- Surface roughness,
- Local property variations due to gradients in slicing process,
- Mechanical properties (Young's modulus, Poisson ration, fatigue stress),
- Conductivity and thermal expansibility.