Autonomous High-Throughput Materials Discovery of Metal Oxide Thin Films via Laser Spike Annealing, Spatially Resolved X-Ray Diffraction, and Thin Film Device Characterization

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Primary CNF Tools Used: CHA Thermal Evaporation, General Photolithography Equipment, Developing Tools, Mask Writers, Wafer Scale Tube Furnaces, Some Metrology Equipment

Abstract:

The study of complex oxides through combinatorial efforts would require a prohibitive amount of material resources and time if experiments were conducted using bulk techniques and at macroscopic length scales. We employ leading edge materials processing in conjunction with advanced micron-scale synchrotron characterization to enable these high-throughput studies and accelerate materials optimization and discovery. This research area is additionally enhanced by incorporating artificial intelligence and machine learning methods. Dimensionality reduction and reproducibility of the processing techniques requires a precision registration of the micron-scale x-ray characterization is essential for analytical autonomous research. We use the Cornell NanoScale and Science and Technology Facilities (CNF) in order to align each of the experimental elements with a primary goal of discovering novel materials. Thin film capacitor devices are constructed post material processing to allow electrical characterization of the properties of the materials synthesized.

Summary of Research:

The focus of the van Dover research group is combinatorial and autonomous materials exploration via thin film synthesis, processing, and characterization. The scale of possible material combinations becomes exponentially large for thin films with a composition spread spanning multiple degrees of chemical freedom, and processing adds even more dimensions of complexity. While exhaustive searching of a system may be impossible, strategically planned experiments can dramatically reduce the resources required to completely map composition/processing space. We collaborate with experts to develop and deploy state of the art artificial intelligence methods to accelerate the discovery of material systems [1,2].

Combinatorial thin film libraries are created by reactively sputtering atoms from one or more targets onto a 4-inch silicon wafer, followed by processing the as-deposited material with a thermal treatment technique called laser spike annealing (LSA) in collaboration with the Thompson group (Figure 1). Co-sputtering in an off-axis geometry with two or more targets produces a monotonic gradient in chemical makeup of the thin film across the

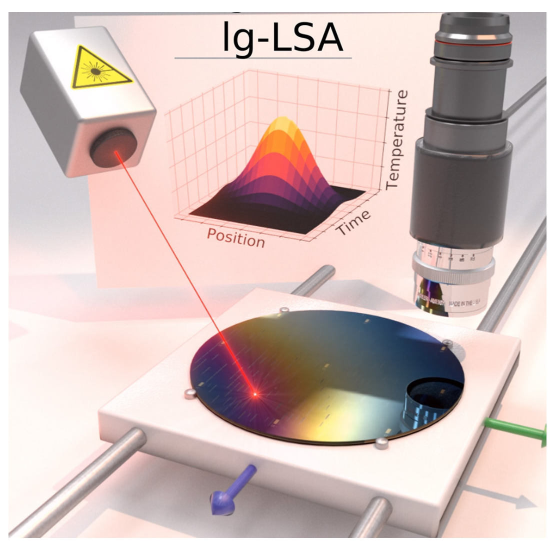


Figure 1: A rendering of the laser spike annealing experiment in relation to the as-deposited thin film and analytical methods.

substrate. Increasing the number of targets decreases the area on the film with unique composition; the trade-off of span in chemical space is the scarcity. LSA controls the peak temperature and duration at that temperature; i.e., the thermal history the material experiences. LSA can be localized to a 2 mm by 5 mm area on the wafer, allowing a regular gird of 616 independently chosen LSA conditions.

There are an additional nine locations photolithographically defined on the wafer that enable precise alignment among various spatially resolved techniques used to analyze the processed materials (e.g., optical microscopy, thin film reflectance spectroscopy, and spatially resolved x-ray diffraction characterization).

Figure 2 shows the layout of the annealed stripes on a composition spread and the total number of conditions for an example material system. To make the experimentation reproducible and precise (especially critical for automation and autonomous learning,) prior to thin film deposition every silicon wafer has the location grid and calibration marks lithographically patterned using CNF.

We successfully processed and characterized (see one example stripe in Figure 3) over 30 unique thin film metal oxide libraries at the Cornell High Energy Synchrotron Source in April 2022. Five of those libraries were used in successful active learning trials yielding a marked improvement in throughput of materials exploration and demonstrating integration of artificial intelligence into experimental materials research. Four of the other libraries were of interest for their electrical properties and further processed into a dense array of thin film capacitor devices which are currently being measured. The remainder of the combinatorial libraries are being analyzed and the most illuminating results are being prepared for publication. An example product from the x-ray characterization is the processing phase diagram in Figure 4.

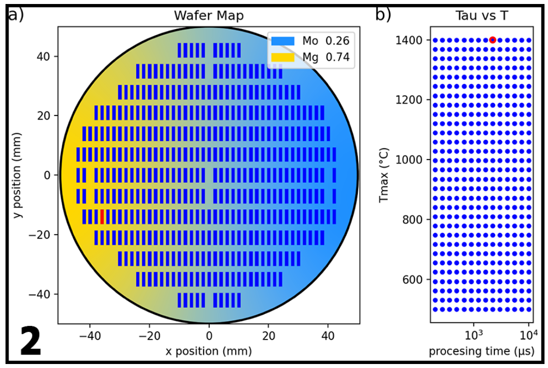
Conclusions and Future Steps:

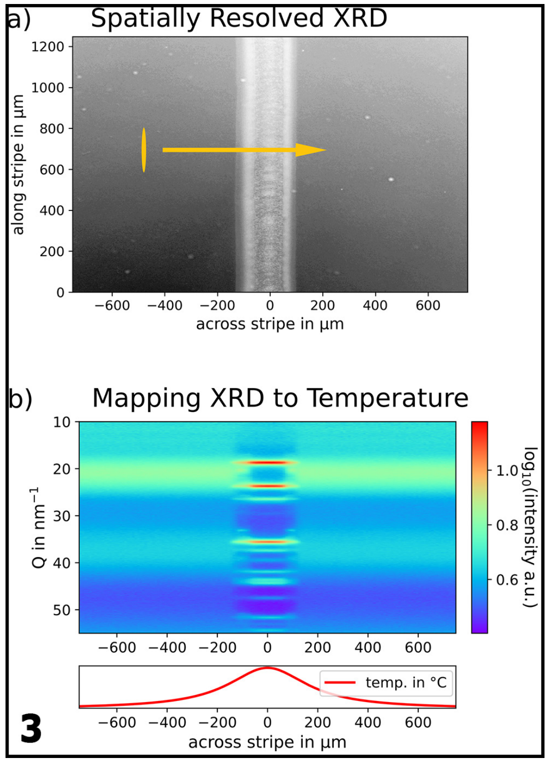
We have engineered an autonomous materials synthesis and characterization system for accelerating the discovery of novel materials. The reproducibility, reliability, and analytical requirements for automating the experimentation and incorporating the artificial intelligence was made possible by utilizing the CNF. Future research efforts will focus on improving the temporal and thermal calibration of the laser spike annealing method using lithographically patterned phase-change materials. One limitation of the post processed capacitor devices is the time required to exhaustively measure the properties each stripe of annealed material. We plan to employ artificial intelligence to optimally measure the minimum number of capacitors required to map structure/property relations by taking advantage of information derived from x-ray characterization. Developing this foundation will allow us to construct more complicated devices for assessing transport and carrier properties of the electronic materials.

References:

[1] Ament, S., et al. Sci. Adv. 7, eabg4930 2021.

[2] Sutherland, D., et al. ACS Combi. Sci. 2020, 22, 12, 887-894.





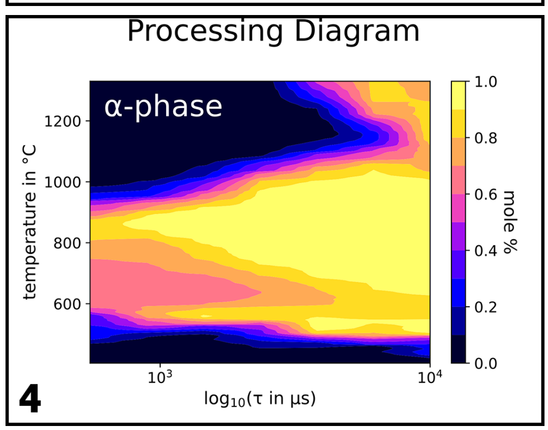


Figure 2, top: a) Cartoon description showing the distribution of laser spike annealing experiments over a 4" wafer with a pseudo binary composition spread of the cations Mg and Mo. b) Distribution of laser spike annealing conditions in the materials processing space. Figure 3, middle: a) Optical micrograph of a stripe of annealed material; yellow spot indicates the footprint and aspect ratio of the x-ray diffraction beam and arrow indicates direction of data collection. b) The spatially resolved x-ray diffraction heatmap comprised of the individual 1D diffraction patterns with the conversion of spatial extent to effective temp of annealing. Figure 4, bottom: A novel quantitative processing phase phase diagram for a laser spike annealed thin film oxide.