Big-Data Approach to Surface Diffraction Data for Film Growth

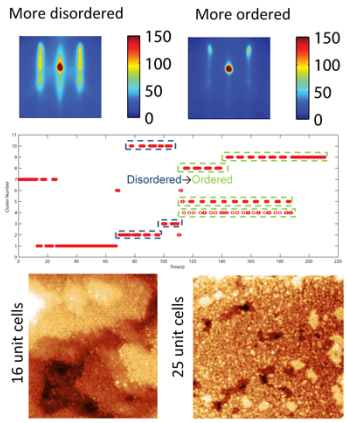

Fig: Top: k-means clustering of RHEED, with two clusters (out of 10) shown above. Time dependence of clusters is shown center. Bottom: Scanning Tunneling Microscopy images of 16 unit cell LCMO and 25 unit cell LCMO, showing the change in growth mode for the thicker film

Work was performed as a user project at the Center for Nanophase Materials Sciences, ORNL

Scientific Achievement

Multivariate statistical analysis is applied to surface diffraction data to unravel hidden epitaxial film growth processes.

Significance and Impact

Full information capture and statistical analysis of reflection high energy electron diffraction (RHEED) data was developed to yield new insight into epitaxial film growth.

Research Details

- RHEED has allowed monitoring of film growth with unit-cell precision in since the 1980s. Yet, the majority of information in RHEED data is not analyzed.
- We use multivariate statistical analysis including principal component analysis, k-means clustering and Fourier analysis and uncover a disorder-order growth transition which is verified by Scanning Tunneling Microscopy in $La_{3/8}Ca_{5/8}MnO_3$ thin films.
- These studies show the utility of Big-Data statistical approaches, providing fundamental understanding and full analysis that will allow to improve film quality.

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Narrative Highlight Text: Reflection High Energy Electron Diffraction (RHEED) is a mainstay of film growth chambers worldwide, allowing in-situ monitoring of films grown by molecular beam epitaxy (MBE) or pulsed laser deposition (PLD). Researchers have used the intensity oscillations of the specular RHEED reflection to determine the layer coverage and thus provide control over thickness. However, there is a wealth of other static and dynamic information contained in the RHEED images, such as spatial extent of ordering (step density), the presence of surface reconstructions, and transitions from two-dimensional to three-dimensional growth. However, due to the large data size of a RHEED image sequence, much of this information is discarded due to lack of appropriate computational tools. Here, we apply multivariate statistical approaches to analyze an entire RHEED movie of $La_{3/8}Ca_{5/8}MnO_3$ growing ion (001) $SrTiO_3$ substrate. The analyses used include Principal Component analysis, which greatly reduces data size and highlights the relevant behaviors in the film, and Fourier analysis, which isolates the transmission spots arising from non-ideal layer by layer growth of the front. K-means clustering reveals the presence of clusters that are largely streaky, but transition to a more spot-like pattern towards the latter stages of the growth, suggesting a transition to a more ordered growth. These results are confirmed through *in-situ* atomically resolved real-space images of LCMO films of varying thicknesses. Overall this study details an effective method to analyze RHEED data in a systematic fashion without information loss, and further could potentially be applied *in-operando* using forward-prediction algorithms to increase epitaxial film quality over current levels.

Reference

"Big-data Reflection High Energy Electron Diffraction Analysis for Understanding Epitaxial Film Growth Processes"

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