Quantifying Defect Pathways for Disorder in La$_{1-x}$Sr$_x$FeO$_3$ / SrTiO$_3$ Thin Films

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Determining the behavior of functional materials under irradiation is important for fundamental understanding of order-disorder phenomena as well as control of performance in extreme conditions for applications such as nuclear reactors or spacecraft. Prior studies of model oxide interfaces have found that certain interface configurations may be more robust to amorphization under irradiation [1]. These findings raise the question of how initial defect distributions with a thin film may affect the evolution of disorder and how such populations can be tuned to guide the radiation response. Here we study the progression of disorder in oxide thin films using analytical scanning transmission electron microscopy, revealing how intrinsic defect populations guide disordering pathways.

To determine how different initial states affect the accumulation of disorder due to ion irradiation, we have examined the material system La$_{1-x}$Sr$_x$FeO$_3$ (LSFO) grown on SrTiO$_3$ (001). Thin films are epitaxially grown by pulsed laser deposition and molecular beam epitaxy, producing largely single crystal films, with some additional LFO films grown containing columnar defect domains. To investigate the different stages of disorder leading to amorphization, films were masked and irradiated to progressively higher doses in quadrants to 0.1, 0.5, and 1 dpa with a 2.8 MeV Au^{2+} ion beam at 3° off the normal to reduce channeling effects. Samples were examined by scanning transmission electron microscopy (STEM) imaging and energy dispersive X-ray spectroscopy (EDS) prior to irradiation to determine film quality and after irradiation to evaluate the effects. Finally, the distribution of crystalline and amorphous phases was measured using few-shot machine learning (ML).

The schematic in Figure 1 portrays the process of the study: samples representing different initial defect configurations are irradiated with ions and are analyzed at intermediate irradiation steps to examine the amount of disorder introduced until full amorphization occurs. The top of the graphic shows STEM high-angle annular dark field (STEM-HAADF) micrographs for single crystal LFO (pristine single crystal film), La$_{0.75}$Sr$_{0.25}$FeO$_3$ (alloyed film), and LFO with defect domains (containing defect domains) prior to irradiation. We observe highly non-uniform pathways for disorder, tied to local compositional fluctuations and intrinsic oxygen vacancy content. We evaluate the observed behavior and propose methods to tune the radiation response of these materials [2].
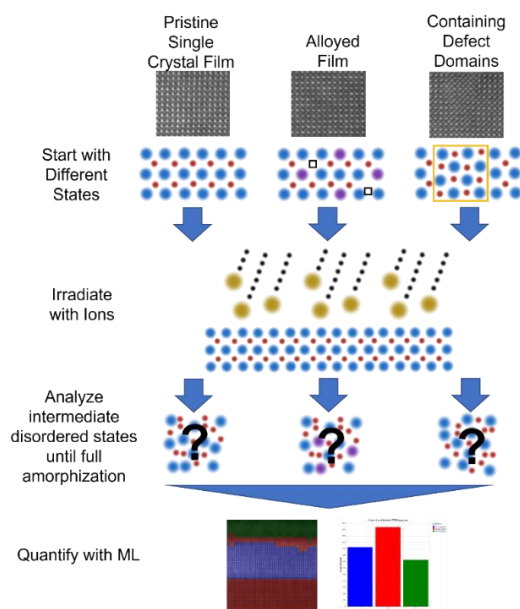
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Figure 1. Schematic overview highlighting different initial defect configurations and quantification of defect-mediated pathways for radiation-induced disorder.

References:

[1] BE Matthews et al., Nano Letters **21**(12) (2021), p. 5353.
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