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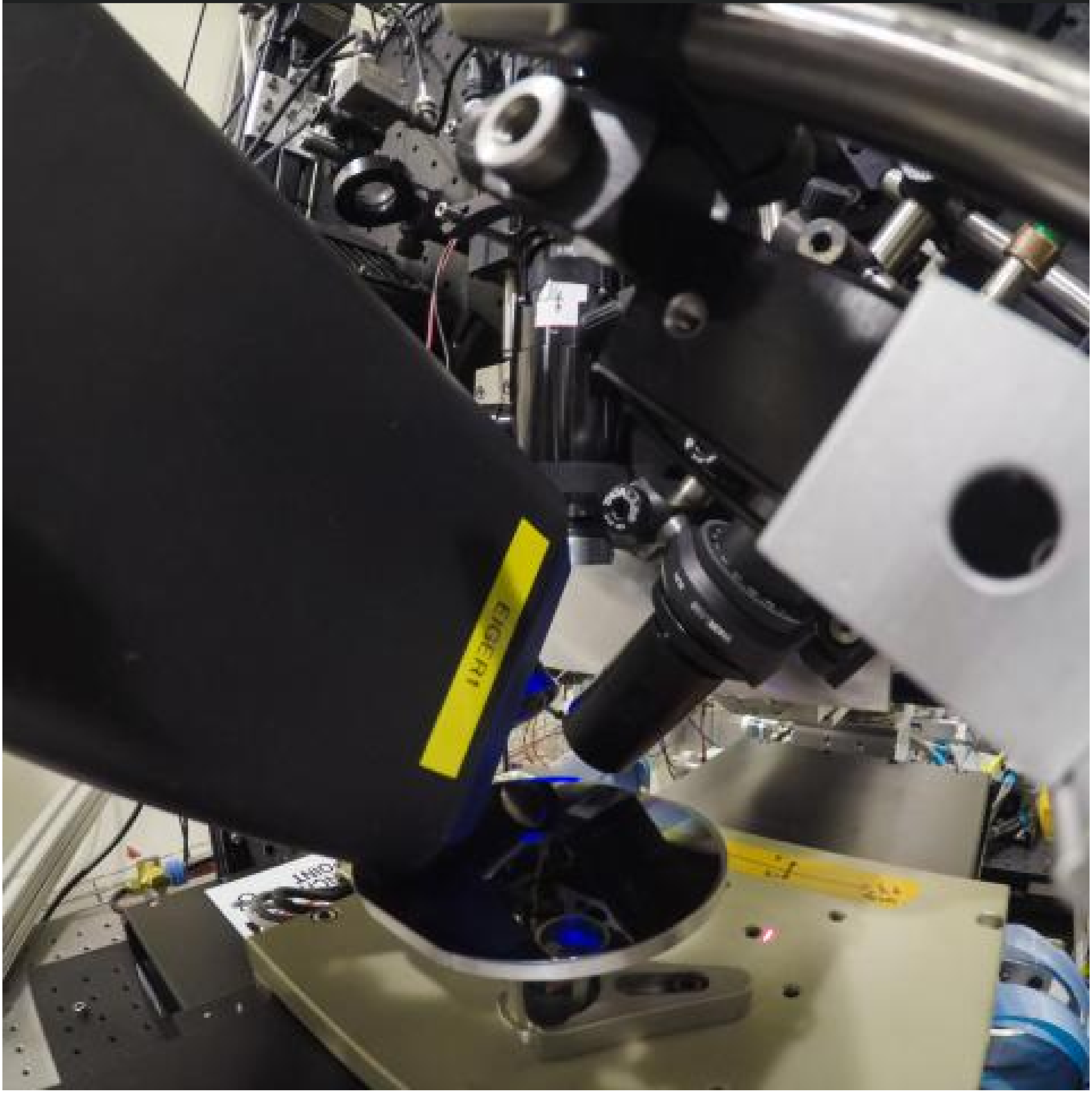
Autonomous materials development using in situ laser annealing and scan-probe, grazing incident x-ray microdiffraction.

April 13, 2021 | D. Sutherland, M. Amsler, S. Ament, Katie R. Gann, A. Connolly, M.-C. Chang, C. P. Gomes, M. O. Thompson, R. B. van Dover

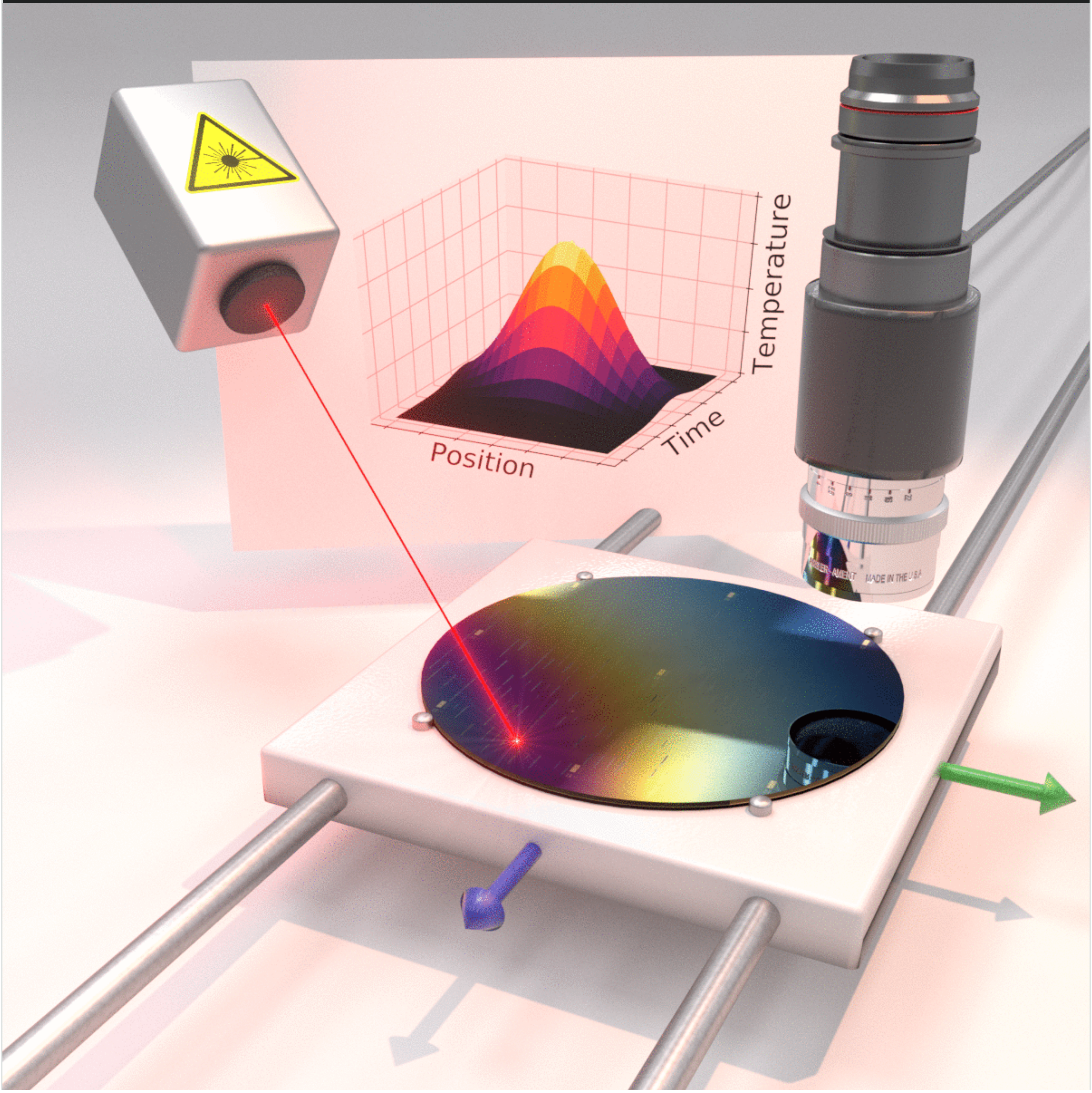
During the 2021-1 run cycle at the FMB-beamline of MSN-C, an interdisciplinary group of researchers based at Cornell University demonstrated the first use of an AI-directed, fully-automated process for thin-film metastable materials exploration.



Automonous laser annealing / GIWAXS setup at the FMB beamline at CHESS.

The experiment employed an AI-driven, fully autonomous infrastructure controlled across campus from CHESS to direct the synthesis and characterization in combinatorial thin-film material processing. Using high-throughput, closed-loop robotics, new materials were synthesized through laser-annealing, analyzed with x-ray diffraction, and the resulting data was fed into a machine-learning model to propose the next best experiment.

Materials **synthesis** was carried out on thin films of various chemical systems by rapidly transiting a high-powered IR laser over a small region of the sample to induce crystallization from the amorphous precursor. Precise control of the IR laser allows selection of distinct quench rates and temperatures, which in turn control the formation of different crystalline phases, including metastable phases not normally present at room temperature. Such metastable materials are extremely challenging to recover to ambient conditions, but once synthesized these phases exhibit unique properties, pushing the boundaries of materials science. Each anneal cycle uses only 0.2% of the available thin-film area, permitting exploration of hundreds of different annealing conditions on a single wafer. We focused our work on oxide materials, which are promising for a wide range of renewable energy applications, such as catalysts and solid oxide fuel cells.

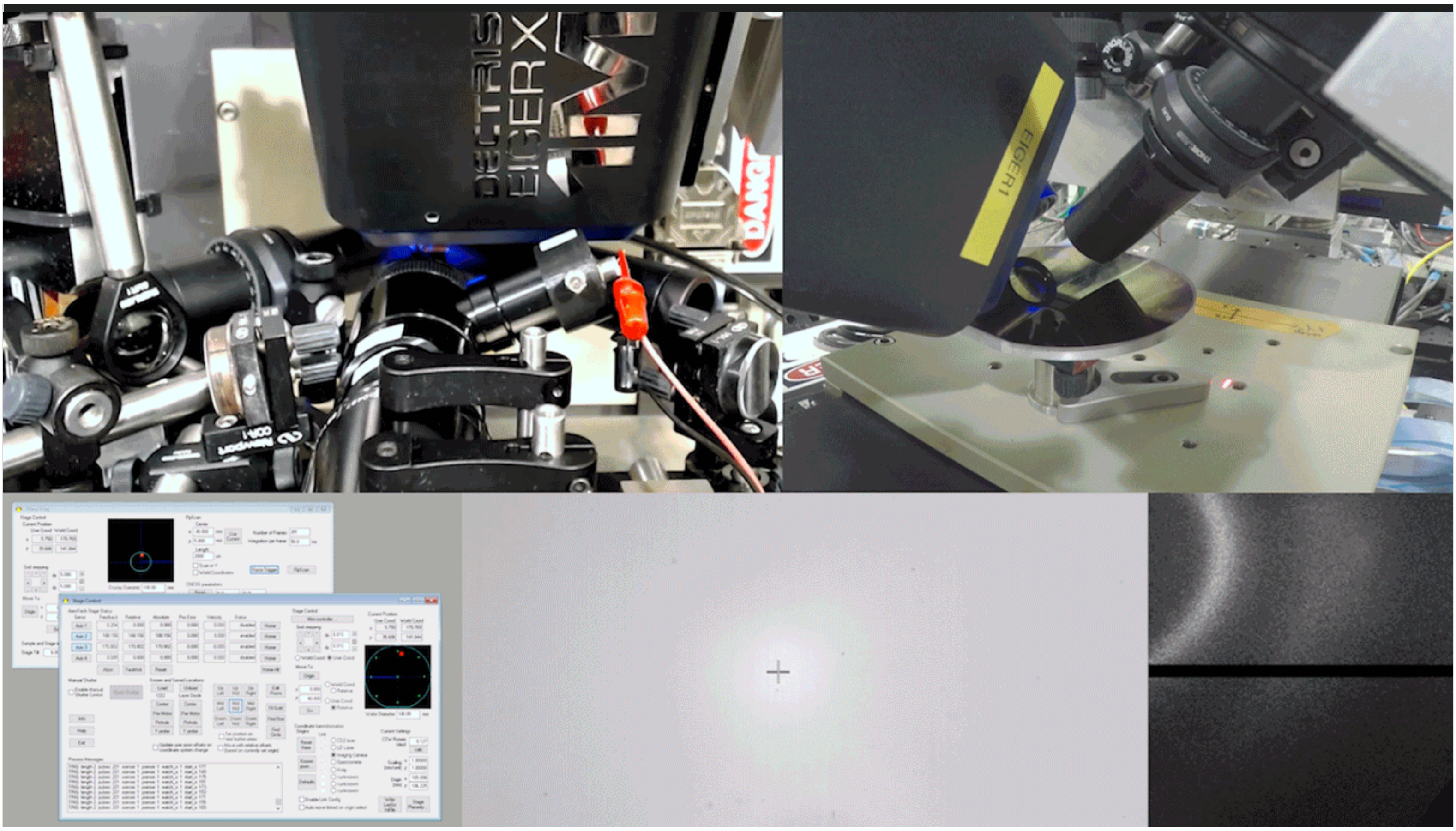


Schematic rendering of the autonomous processing-characterization loop via optical feedback. The IR laser transits across a region of the film, achieving the desired processing temperature and time/quench rate. The processed stripe is analyzed via optical and microfocused x-ray diffraction (not shown here) which is fed back into the AI to inform the next best laser experiment.

To **analyze** the crystalline nature of the laser-annealed films, microfocused x-rays were necessary due to the small experimental footprint. The beam was scanned across the anneal stripes to capture the intricate changes in the diffraction patterns at a high spatial resolution. The results of those scans, each comprising around 200 x-ray diffraction images, were then processed on-the-fly and fed into a machine-learning model for further analysis.

Finally, an AI-driven software used this information to iteratively decide which experiment to perform next. This machine-learning scheme, also referred to as “active learning”, aims at minimizing the number of experiments to thoroughly and efficiently explore the space of metastable materials.

At the heart of these experiments is the scientific autonomous reasoning agent, nicknamed “SARA: The Robot Scientist”. SARA orchestrates the execution of the synthesis robotics, collects the CHESS x-ray diffraction data, and queries the AI for the next experimental conditions. Following the previous CHESS experiment in December 2019, the general approach of the complete autonomous closed-loop setup was simulated post facto in preparation of the current beamtime, and allowed us to benchmark and optimize our implementation of SARA to determine the most effective approach. In the 2021-1 run cycle at the FMB-beamline, we were finally able to put our implementation of SARA to the test for the first time in a real-time experiment, and were able to demonstrate a closed-loop materials exploration cycle.



A video demonstrating a few iterations of the fully autonomous closed-loop active learning from the 2021-1 CHESS run. “SARA” orchestrated every aspect of the experiment, from the processing hardware (panels a-c IR laser, optical imaging camera, Dectris Eiger 1M detector, and the LasGo translation stage software) to characterization of the annealed film (panels d and e showing the microscope imaging and x-ray detector output), learning the behavior of the material under highly non-equilibrium processing conditions.

What are the broader impacts of this work?

Accelerating the pace of **materials discovery/development** will enable the deployment of new technologies. The integration of AI in scientific experimentation will dramatically accelerate materials explorations, especially with high-throughput methods to generate data to advance our understanding of new materials. The methods developed here can be applied to various materials classes, leading to the design of better fuel cells, photocatalysts, and battery materials

Why is this important?

This work is a major step towards autonomous experimentation in which an AI system not only directs automated characterization of materials, but also uses the resulting data to inform the next automated synthesis experiment.

Why did this research need MSN-C & CHESS?

The X-ray experiments in this study were done exclusively at CHESS. The ID3B beamline offers the ability to direct a microfocussed (10 μm x 3 μm) high-intensity beam to interrogate oxide samples mounted on a custom high-speed x-y stage, enabling spatially-resolved characterization of many thousands of synthesis conditions and compositions per hour.

How was the work funded?

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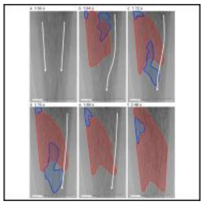

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