Scientists track patterns of island growth in crystals

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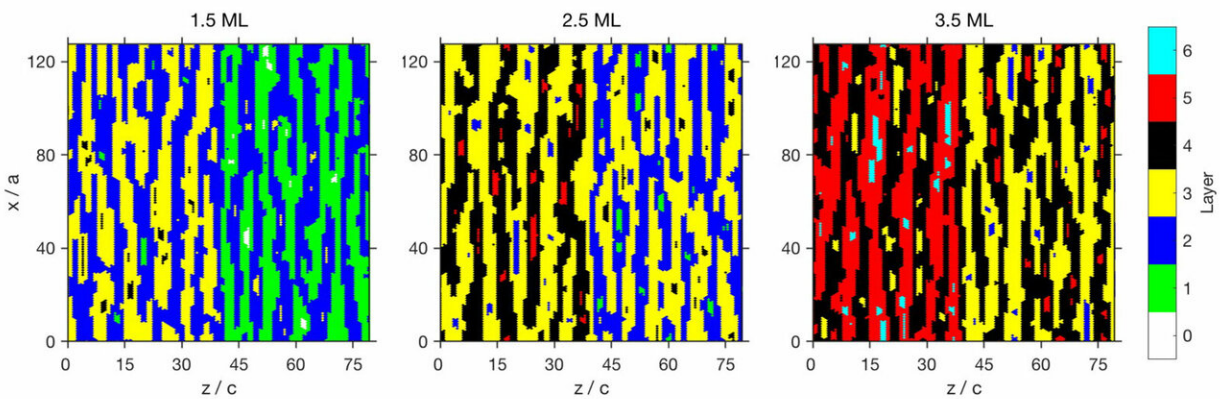

This image from a simulation shows the formation of islands during layer-by-layer growth of a gallium nitride crystal. In this image, each color corresponds to a different layer and snapshots are shown at different points in time. A new discovery has shown that each layer tends to form in a pattern similar to the preceding layer. Credit: Argonne National Laboratory

Argonne scientists reveal connections as crystalline layers form.

Understanding how crystals grow impacts broad areas of materials science, from developing better microelectronics to discovering new materials. At the atomic level, crystals can grow in several different ways, and scientists have recently discovered an intriguing behavior associated with a common way that crystals grow.

In this mode of crystal growth, called ""layer-by-layer," the surface of the crystal starts out very smooth at the atomic level. New atoms that arrive on the surface tend to skate around until they find each other. When this happens, they begin to form a new one-atom-thick layer by joining, creating a flat region known as an island. As more atoms arrive, additional islands form at other places on the surface. Eventually the growing islands cover the whole surface, coalescing to form a new atomic layer.

"If we understand how crystals grow in this mode, we might be able to better understand some of the mechanisms behind defect formation, as well as develop techniques to synthesize new types of crystals," said Peter Zapol, Argonne materials scientist.

In a new study from the U.S. Department of Energy's (DOE) Argonne National Laboratory, scientists have found that the seemingly random arrangement of islands that form to begin new layers can actually be very similar from layer to layer.

Using coherent X-ray scattering techniques to observe the crystal surface at the atomic scale during crystal growth, the researchers were able to characterize the exact arrangements of the islands as they form, or ?"nucleate," in each layer of the crystal.

"You can think of what we're doing as something like making pancakes in a pan," said Argonne Distinguished Fellow and study author Brian Stephenson. ?"As we randomly add more atomic ?'batter,' our pancake islands start to run together and coalesce. The interesting thing is that every time we grow a new layer, the pattern of pancakes repeats the pattern of the original layer."

One important consideration that Stephenson noted is that the nucleation of new islands was not influenced by defects in the crystal structure—which is to say, it was not controlled by static regions where nucleation would be most likely to occur.

"This is a dynamic relationship; the layer that is almost completely grown communicates with the layer that is beginning to grow on top of it," said Argonne physicist Peter Zapol, another author of the study.

As the lower layer continues to fill in, the remaining holes tend to occur in areas far away from the original nucleation sites. Because these holes discourage next-layer nucleation in their vicinity, nucleation of the next layer will tend to take place far away from the holes and close to the original nucleation sites.

"The persistent patterns that we see indicate that there is communication between the layers," Stephenson said. ?"There is a vestige of the first layer that gives information to the next one."

The ability to characterize the island patterns comes as a result of the researchers' use of coherent X-rays provided by Argonne's Advanced Photon Source, a DOE Office of Science User Facility. According to Stephenson, incoherent X-rays used in previous experiments were able to reveal only average features of the island landscape, while coherent beams are sensitive to the exact island arrangement.

"The old way just told us the average spacing and shape of the islands—with coherent X-ray beams, we're able to generate a whole lot more information," he said. ?"The resolution has gotten so good that we're now able to resolve correlations across the whole sample, which means we can see things like this pattern that tell us how the islands relate to each other."

Modeling growth dynamics at the atomic level helped the researchers achieve a deeper understanding of crystal growth, Zapol said. ?"If we understand how crystals grow in this mode, we might be able to better understand some of the mechanisms behind defect formation, as well as develop techniques to synthesize new types of crystals."

A paper based on the study, "Coherent X-ray spectroscopy reveals the persistence of island arrangements during layer-by-layer growth," appeared in the March 4 issue of *Nature Physics*. Other Argonne authors included Guangxu Ju, Dongwei Xu, Matthew Highland, Jeffrey Eastman, Paul Fuoss, and Hua Zhou. Carol Thompson from Northern Illinois University and Hyunjung Kim from Sogang University in South Korea also contributed.

More information: Guangxu Ju et al, Coherent X-ray spectroscopy reveals the persistence of island arrangements during layer-by-layer growth, *Nature Physics* (2019). DOI: 10.1038/s41567-019-0448-1

Provided by Argonne National Laboratory

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