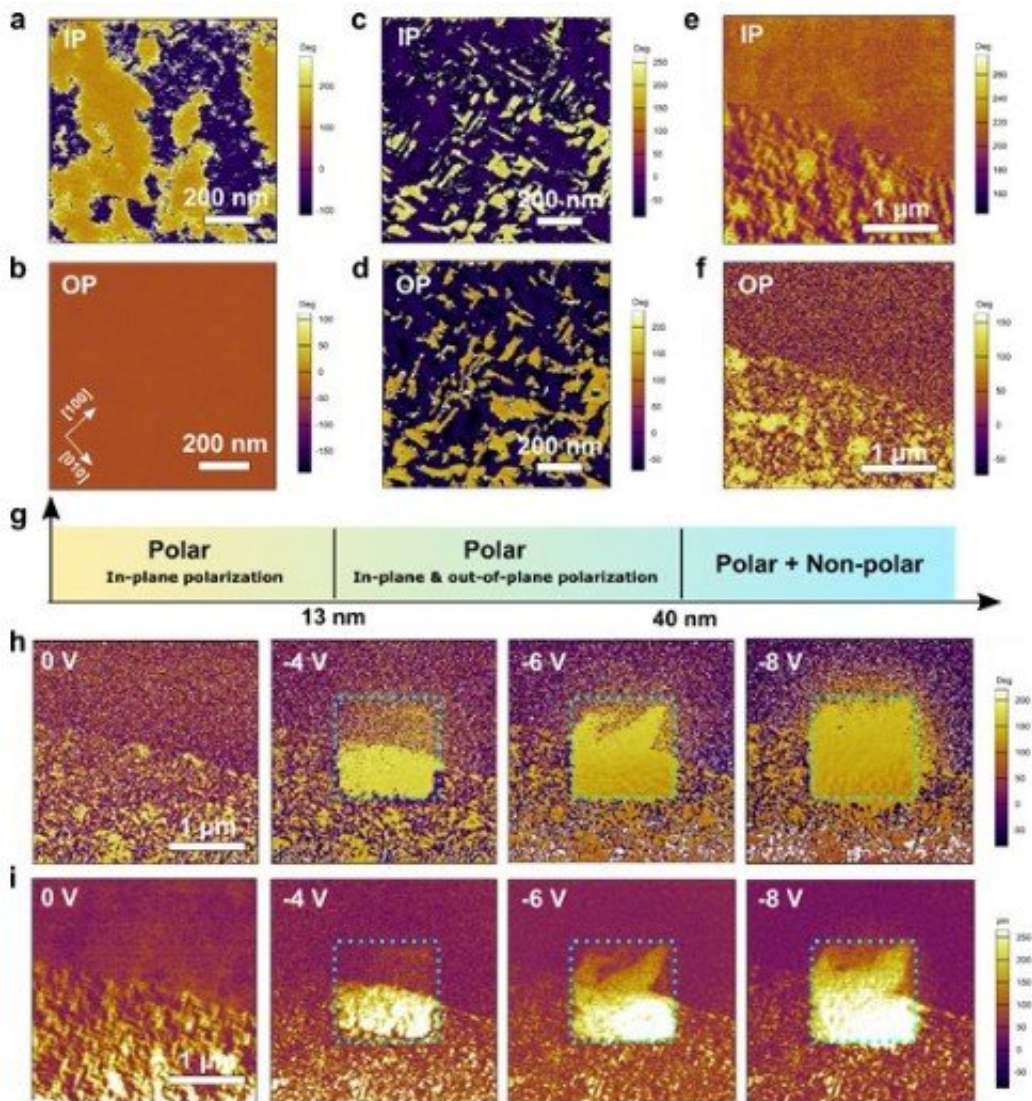


Make them thin enough, and antiferroelectric materials become ferroelectric

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Piezoresponse force microscopy (PFM) of emergent ferroelectricity and phase coexistence in NaNbO_3 membranes. Lateral and vertical PFM phase images obtained for a) and b) 13 nm, c) and d) 40 nm, and e) and f) 100 nm thick

membranes, respectively. g) Schematic phase diagram summarizing the observed ferroic orders in three thickness regimes of NaNbO_3 membranes probed by PFM. Vertical h) phase and i) amplitude PFM images measured near the mixed phase region in the 100 nm thick membranes after applying a tip bias of 0V, -4V, -6V, and -8V. Credit: *Advanced Materials* (2023). DOI: 10.1002/adma.202210562

Antiferroelectric materials have electrical properties that make them advantageous for use in high-density energy storage applications. Researchers have now discovered a size threshold beyond which antiferroelectrics lose those properties, becoming ferroelectric.

"Electronic devices are getting smaller and smaller, which makes it increasingly important for us to understand how a material's properties may change at small scales," says Ruijuan Xu, corresponding author of a paper on the work and an assistant professor of materials science and engineering at North Carolina State University.

"In this case, we learned that when antiferroelectric thin films get too thin, these materials go through a phase transition and become ferroelectric. That makes them less useful for energy storage, but creates some new application possibilities for memory storage."

This research focused on antiferroelectric materials. These materials have a [crystalline structure](#), which means they consist of regularly repeating units. Each repeating unit in the crystalline structure has a "dipole"—a positive charge paired with a negative charge. What makes antiferroelectric materials special is that those dipoles alternate from unit to unit throughout the structure.

In other words, if one unit has a positive charge on "top" and a negative

charge on the "bottom," then the next unit will have the [positive charge](#) on the "bottom" and the [negative charge](#) on "top." This regular spacing of the dipoles also means that, at the macroscale, antiferroelectric materials have no positive or negative polarization.

Ferroelectric materials also have a crystalline structure. But in ferroelectrics, the dipoles in the repeating units all point the same way. What's more, you can reverse the polarization of the dipoles in [ferroelectric materials](#) by applying an electric field.

To explore how an antiferroelectric material's properties may change at small scales, the researchers focused on lead-free sodium niobate (NaNbO_3) membranes.

Antiferroelectric thin films are grown on a substrate. Previous attempts to assess potential size-related effects on antiferroelectric thin films have looked at the thin films while the films are still attached to the substrate layer. This poses significant challenges, because there are "strains" where the thin film is strongly connected to the substrate—and it is difficult to assess what effects are related to the thin film's size and what effects are caused by the strains related to the substrate.

"To address this challenge, we introduced a sacrificial buffer layer between the antiferroelectric thin film and the substrate," Xu says. "Once we had grown the thin film to the desired thickness, we selectively etched the sacrificial layer. This allowed us to detach the thin film from the substrate. Ultimately, this allowed us to determine how any changes in the thin film are affected by its size, because we knew the substrate was not contributing to any changes."

The researchers then used a variety of experimental and theoretical approaches to assess these strain-free samples at thicknesses ranging from 9 nanometers (nm) to 164 nm.

"The results were quite unexpected," Xu says.

"We know that at the atomic scale, antiferroelectric materials—like lead-free NaNbO_3 membranes—have alternating dipoles throughout the material. We found that when the NaNbO_3 membranes were thinner than 40 nm, they become completely ferroelectric. And from 40 nm to 164 nm, we found that the material had some regions that were ferroelectric, while other regions were antiferroelectric."

Using their [experimental data](#), the researchers extrapolated there would be at least some ferroelectric regions in the NaNbO_3 at any thickness below 270 nm.

"One of the exciting things we found was that when the [thin films](#) were in the range where there were both ferroelectric and antiferroelectric regions, we could make the antiferroelectric regions ferroelectric by applying an electric field," Xu says. "And this change was not reversible. In other words, we could make the thin film completely ferroelectric at thicknesses of up to 164 nm."

The researchers were also able to draw some conclusions on what is driving these changes in the antiferroelectric material.

"Drawing on first principles, we were able to conclude that the phase changes we see in exceptionally thin antiferroelectric materials are driven by structural distortion that begins on the membrane's surface," Xu says.

In other words, instabilities at the surface have a ripple effect that runs throughout the material—which isn't possible when the volume of the material is higher. That's what prevents antiferroelectric materials from becoming ferroelectric at larger scales.

"I don't want to speculate too much about potential applications, but our work offers significant insights into how we can control a material's properties by taking advantage of size effects," Xu says. "We've demonstrated significant size effects in NaNbO_3 , and the techniques we used to uncover those effects can be used to explore similar questions for a range of other materials."

The paper, "Size-Induced Ferroelectricity in Antiferroelectric Oxide Membranes," is published open access in the journal *Advanced Materials*.

More information: Ruijuan Xu et al, Size-Induced Ferroelectricity in Antiferroelectric Oxide Membranes, *Advanced Materials* (2023). [DOI: 10.1002/adma.202210562](https://doi.org/10.1002/adma.202210562)

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