Two-dimensional limit of crystalline order in perovskite membrane films

Seung Sae Hong,1,2,3# Jung Ho Yu,2 Di Lu,2,4 Ann F. Marshall,5 Yasuyuki Hikita,3 Yi Cui,2,3,6 Harold Y. Hwang1,2,3#

Long-range order and phase transitions in two-dimensional (2D) systems—such as magnetism, superconductivity, and crystallinity—have been important research topics for decades. The issue of 2D crystalline order has reemerged recently, with the development of exfoliated atomic crystals. Understanding the dimensional limit of crystalline phases, with different types of bonding and synthetic techniques, is at the foundation of low-dimensional materials design. We study ultrathin membranes of SrTiO3, an archetypal perovskite oxide with isotropic (3D) bonding. Atomically controlled membranes are released after synthesis by dissolving an underlying epitaxial layer. Although all unreleased films are initially single-crystalline, the SrTiO3 membrane lattice collapses below a critical thickness (5 unit cells). This crossover from algebraic to exponential decay of the crystalline coherence length is analogous to the 2D topological Berezinskii-Kosterlitz-Thouless (BKT) transition. The transition is likely driven by chemical bond breaking at the 2D layer-3D bulk interface, defining an effective dimensional phase boundary for coherent crystalline lattices.

INTRODUCTION

Arguments based on statistical mechanics (1) rigorously excluded the spontaneous breaking of continuous symmetries in two-dimensional (2D) systems, indicating the absence of long-range order (Mermin-Wagner theorem). However, it was later suggested that 2D systems can maintain a quasi-ordered phase at low temperature if they can host pairs of topological defects—such as vortex or dislocation pairs—which maintain continuous symmetry with finite energy (2–9). The unpairing of these topological defect pairs can collapse quasi-long-range order and introduce an unusual phase transition known as the Berezinskii-Kosterlitz-Thouless (BKT) transition, which was experimentally shown in a few systems (10–13). 2D crystalline order is one of the 2D systems initially predicted to undergo the BKT transition by having edge dislocation pairs as the topological defects (3, 5, 6, 9). However, the experimental study of the BKT transition in crystalline lattices has been rare, because the thermal fluctuation energy is typically hundreds of times smaller than chemical bonding energies in crystalline materials, limiting experimental studies to artificial 2D crystalline systems, such as colloidal spheres (14). Within this historical context, the discovery of graphene and its high-crystalline quality are extraordinary (15). It is now understood that graphene forms intrinsic ripples to stabilize the lattice without violating the Mermin-Wagner theorem (16–18). Although graphene and other atomic crystals represent examples of 2D crystalline phases without apparent phase transitions on accessible sample length scales, it is an open question to determine the general phase diagram for 2D lattices, with different chemical properties and bonding nature.

RESULTS

Structural characterization of ultrathin SrTiO3 membranes

SrTiO3 is a perovskite oxide compound with ionic bonding and has a cubic structure at room temperature. SrTiO3 is known for its wide physical and chemical stability windows; for example, the SrTiO3 (001) surface does not undergo reconstructions for moderate temperatures (19). The TiO2-terminated surface is stable in most common solvents; thus, SrTiO3 is a popular substrate for oxide thin film growth (20). The stability and the simple crystal structure suggest that SrTiO3 is an ideal crystalline lattice for studying 2D lattice physics without dominant contributions from chemical reactions and thermal decomposition. Here, we study the structural evolution of SrTiO3 in the 2D limit, taking advantage of recently developed methods for fabricating freestanding thin films (21, 22). SrTiO3 crystalline membranes of different thicknesses were synthesized by pulse laser deposition (PLD), as previously reported for synthesis of large-area freestanding oxide layers (21). A water-soluble sacrificial layer (Sr3Al2O6) and the oxide film of interest (SrTiO3) were grown epitaxially on a SrTiO3 (001) substrate (Fig. 1A), and the sacrificial layer was selectively dissolved at room temperature in deionized water to release the top SrTiO3 layer. Both in situ and ex situ characterization confirmed that the oxide layers grown on the substrate were macroscopically single-crystalline, for which thicknesses were controlled with single u.c. (0.39 nm) accuracy (see Materials and Methods).

We examined the crystalline structure of freestanding SrTiO3 membranes (Fig. 1B) using TEM. Figure 1C shows HR-TEM images of SrTiO3 membranes of three different thicknesses (d). In the cases of relatively thicker membranes (8 u.c.), we found that the membrane is effectively single-crystalline with characteristic cubic lattice fringes. However, below a certain thickness, the film evolved to a mixture of crystalline and amorphous domains (4 and 3 u.c.) and further to an almost completely amorphous layer (2 u.c.). To quantify the observed crystalline phase transition, we used selected-area electron diffraction (SAED) patterns and dark-field TEM (DF-TEM) (23). SAED patterns of the entire suspended membrane (Fig. 2A) shows that the orientation of crystalline domains was mostly aligned within the 2-μm-wide area, even for the thinnest (2 u.c.) sample. DF-TEM, a diffraction-filtered imaging tool

#Corresponding author. Email: seungsaehong@stanford.edu (S.S.H.); hyhwang@stanford.edu (H.Y.H.)
using the SAED pattern, was used for spatially mapping the crystalline domains in the membranes of different thicknesses (Fig. 2B).

There are two primary observations from the DF-TEM images. First, the membrane changed from approximately single-crystalline (white) to amorphous (black) as it got thinner, for which domain sizes were well matched to the HR-TEM images. Second, even for single-crystalline membranes, there was a thickness-dependent population of dislocations (local black areas). The density of dislocations observed in the membranes was much larger (~100 times) than the dislocation density originating from the substrate and thin film growth (21, 24, 25). We calculated spatial autocorrelation functions (26) for each thickness and estimated the crystalline coherence length \( L \) to quantify the crystalline order based on DF-TEM images (see Materials and Methods). Figure 2C shows the observed continuous reduction of the crystalline coherence length, with a qualitative change in trend near 5-u.c. thickness.

The observed crystalline to amorphous transition in the 2D limit is surprising, because all SrTiO\(_3\) layers were single-crystalline before release from the bulk substrates (Fig. 3). Several possible extrinsic factors to induce this transition—such as electron beam damage, surface chemical reactions, mechanical strain, and thermal effects (27)—were considered and examined in control experiments (see the Supplementary Materials). They show that SrTiO\(_3\) layers of a few u.c. thickness remain single-crystalline in all circumstances as long as the layer is epitaxially connected to a heterostructure on a bulk substrate, excluding dominant chemical reactions on the SrTiO\(_3\) surface. Instead, the crystalline phase transition occurs only during lift-off when the SrTiO\(_3\) membranes were detached, as confirmed by the surface crystalline structure observed by reflection high-energy electron diffraction (RHEED) in Fig. 3. The lift-off process involves bond breaking at the bottom interface via dissolution of the sacrificial layer \((\text{Sr}_3\text{Al}_2\text{O}_6)\), which releases a large amount of free energy before the broken bonds are passivated (28). This free energy, as large as a few electron volts for ionic bonds, appears to be the main factor driving the structural transition. By contrast, other fluctuation sources—such as relevant mechanical and thermal energies—are orders of magnitude smaller and thus negligible, consistent with the control experiments showing no effect of temperature up to 1150 K and several percent strains (figs. S4 and S5).

**BKT transition in the perovskite oxide membranes**

Here, the structural transition shares much phenomenological similarity with the BKT transition expected for 2D lattices provided that the thickness can be understood to vary the effective “temperature” of the membrane. The BKT transition describes two phases separated by a critical temperature \( T_{\text{BKT}} \) (3, 6, 7): a quasi-long-range–ordered topological phase and a disordered phase. At low temperature \( T < T_{\text{BKT}} \), an effectively 2D crystalline lattice is stabilized with paired edge dislocations, the density of which increases algebraically with increasing temperature. At \( T_{\text{BKT}} \), where thermal fluctuations reach a fraction of the 2D dislocation energy \( E_{2D} \), dislocations unbind and freely proliferate, resulting in the exponential decay of the crystalline coherence length. Both of these phases were observed in the SrTiO\(_3\) membranes of different thicknesses (Fig. 2B). Although the thermal fluctuation scale
As grown:

1 u.c. 2 u.c. 4 u.c. 8 u.c.

Exposed to solvents:

2 u.c. 4 u.c. 8 u.c.

Lifted off from bulk substrates:

No pattern identified

2 u.c. 4 u.c. 8 u.c.

**Fig. 3.** RHEED patterns of SrTiO₃ films before and after solvent exposure and transfer. Top: RHEED patterns of SrTiO₃ films of different thicknesses grown on a Sr₃Al₂O₆ 20-nm film on a SrTiO₃ (001) substrate. Middle: RHEED patterns of SrTiO₃ films of different thicknesses grown on a Sr₃Al₂O₆ 5-nm film on a SrTiO₃ (001) substrate after exposure to all solvents and polymer coatings used in the actual transfer process. The sacrificial layer (Sr₃Al₂O₆) was too thin to be dissolved despite the long immersion in water; therefore, the SrTiO₃ films are still epitaxially integrated to the 3D bulk crystalline heterostructure. Bottom: RHEED patterns of transferred SrTiO₃ membranes (on Si/SiO₂ substrates) of different thicknesses. The RHEED intensity of the 4-u.c. membrane diminished drastically and showed little signature of the crystalline phase. By contrast, the 8-u.c. membrane showed a similar RHEED pattern as the original film. No RHEED pattern was detected from the transferred 2-u.c. membrane.

\( k_B T \sim 25 \text{ meV} \) is far below \( E_{2D} \) even for the thinnest SrTiO₃ membrane, electron volt–level fluctuation energy from bond breaking/passivation processes can be as large as \( E_{2D} \) in membranes of a few u.c. thickness, driving a BKT-like transition (Fig. 4A). For a given fluctuation energy and Young’s modulus, thickness becomes the effective tuning parameter instead of temperature here, with the phase boundary defined by the critical thickness \( d_{BKT} \).

On the basis of continuum elastic theory and the BKT transition model, we calculated the critical thickness \( d_{BKT} \) for SrTiO₃ membranes. A simplified atomic potential model was used to estimate \( E_{2D} \) (deduced from the in-plane bonding potential \( E_i \)) and the chemical fluctuation energy (out-of-plane bonding potential \( E_p \)) equivalent to \( k_B T_{BKT} \). For an ionic atomic potential (Born-Lande model), \( d_{BKT} \) can be expressed as

\[
d_{BKT} = \frac{16\pi E_i}{\alpha E_p a}
\]

where \( \alpha \) is the Born exponent of the lattice and \( a \) is the lattice constant (details are presented in the Supplementary Materials). In SrTiO₃, where \( \alpha = 8 \) (29), for \( E_i = E_p \) (isotropic bonding), \( d_{BKT} \) is 6.3 u.c.

The snapshots of the evolution of crystalline coherence taken by DF-TEM (Fig. 2) enable quantitative analysis and provide compelling evidence for the transition based on BKT physics. We first examine the dislocation size in SrTiO₃ membranes starting from the thick regime. Here, we found that nearly all dislocations are of zero Burgers vector configuration, as expected for paired dislocations (Fig. 4B, inset). The size was almost constant for 6-u.c. membranes and thicker membranes but increased drastically in 5- and 4-u.c. membranes (Fig. 4B). The size plot does not follow the percolation scaling law (30), which excludes the clustering of dislocations in a random manner. The abrupt size change from locally constrained dislocations (6 u.c.) to larger amorphous domains (5 u.c.) strongly suggests that there exists a critical thickness for a dislocation depairing transition between 5 and 6 u.c., close to the estimate from the atomic potential model \( d_{BKT} = 6.3 \text{ u.c.} \) for isotropic bonding—note that the weak bonding anisotropy expected between SrTiO₃ and Sr₃Al₂O₆ would reduce this value. Another important indication of the BKT transition is the exponential scaling observed in the disordered phase. Figure 4C plots the crystalline coherence length \( L \) as a function of the reduced thickness term \( (d_{BKT}/d - 1)^{\nu} \), analogous to the reduced temperature term \( t = (T/T_{BKT} - 1)^{-\nu} \) in the original BKT theory. The critical exponent \( \nu \) from the experimental data is \( \nu = -0.39 \) (Fig. 4C, inset), well matched to the theoretically predicted value \( \nu = -0.4 \) (3, 5, 6). Below the critical thickness, the coherence length is exponentially proportional to the reduced thickness term, which was also observed in other 2D phase transitions (11, 31). The exponential dependence is the characteristic scaling law of so-called infinite-order phase transitions, distinguishing the transition from second-order phase transitions with algebraic scaling.

**General phase diagram of 2D crystalline lattices**

The simple 2D phase transition model introduced here can be expanded to arbitrary crystalline lattices. Figure 5 describes the phase diagram of crystalline order for different thicknesses and bonding anisotropies...
Thus far, many 2D layers have been synthesized in a top-down approach by extraction from 3D crystals (5, 6, 9), identical to L in the disordered phase (d < d_{BKT}) and infinite in the thicker membranes (d > d_{BKT}). In materials with strong bonding anisotropy, interface bond breaking is a minimal energetic perturbation to the 2D crystal below the threshold to induce the phase transition. This corresponds to the case of atomic crystals exfoliated from layered compounds. Graphene and other layered materials have bonding anisotropy as large as several hundred, which excludes a visible crystalline phase transition in the exfoliated crystals and enables high crystalline quality down to the monolayer limit for all practical length scales (15, 32, 33). However, as \( E_{i} \) approaches \( E_{j} \), interface bond breaking can provide sufficient fluctuation energy to drive the transition (Fig. 5). The most extreme case will be the isotropically bonded lattice. Although this phase diagram is based on a specific ionic bonding model, the general trend in the crystalline order versus bonding anisotropy will be similar for different types of chemical bonding.

**DISCUSSION**

SrTiO$_3$ membranes in the 2D limit present a unique experimental crystalline-amorphous system with a phase transition well described by BKT physics. This specific example also suggests the more general relevance of interface chemical energy for the stability of 2D crystalline order, providing a crucial guideline for the synthesis of 2D materials. Weak out-of-plane bonding, membranes derived from 3D crystalline lattices have a universal bound defined by the 2D-3D interface chemistry. This calls for new principles for designing chemical perturbations (for example, artificial interfaces and catalytic bond breaking). Understanding this intrinsic phase diagram of 2D crystalline order will open new opportunities for 2D materials of arbitrary crystalline structures, with controlled crystallinity.

**MATERIALS AND METHODS**

**Sample preparation**

SrTiO$_3$/Sr$_3$Al$_2$O$_6$ films were grown by PLD. On TiO$_2$-terminated SrTiO$_3$ (001) substrates, a 20-nm-thick Sr$_3$Al$_2$O$_6$ film was grown in an Ar partial pressure of \( 5 \times 10^{-6} \) torr with a laser fluence of 1.3 J/cm$^2$, followed by SrTiO$_3$ film growth at an O$_2$ partial pressure of \( 5 \times 10^{-6} \) torr with a laser fluence of 0.6 J/cm$^2$. The substrate was preannealed at 950°C with an oxygen phase pressure of \( 5 \times 10^{-6} \) torr for 30 min, and the growth temperature was maintained at 720°C for both Sr$_3$Al$_2$O$_6$ and SrTiO$_3$ layers. The entire film was grown in a layer-by-layer growth mode, for which the thickness was monitored by RHEED oscillations and later confirmed by atomic force microscopy (AFM) (Fig. S1).

SrTiO$_3$ membranes were transferred to TEM grids by the polymer-transfer method similar to the study of Huang et al. (Fig. S2) (23). A thin layer (200 nm) of polymethyl methacrylate was spun onto the film and baked at 90°C, followed by additional application of polypropylene carbonate (>100 µm) as a mechanical support to prevent bending of membranes. The entire sample was immersed in deionized water at room temperature until the sacrificial layer of Sr$_3$Al$_2$O$_6$ was completely dissolved, and the polymer-SrTiO$_3$ layer was detached from the bulk substrate. The polymer-SrTiO$_3$ layer was scooped by a silicon nitride TEM grid. The polymer support was removed by either thermal decomposition in a tube furnace heated at 250°C in flowing O$_2$ or acetone wet etching followed by a critical point drying step. Both thermal decomposition and wet etching gave identical results for SrTiO$_3$ membranes of the same thickness.

**TEM image acquisition and image analysis**

HR-TEM and DF-TEM experiments were conducted using FEI Tecnai G2 F20 X-TWIN TEM and FEI Titan 80-300 TEM, operated at either 80 or 300 kV. Acquisition times for DF-TEM images were 3 to 10 s. The spatial resolution of DF-TEM imaging is 1 nm. A Gatan 628-0500 in situ heating holder system was used to increase the sample temperature up to 900°C. All DF-TEM images were taken at the field of view where no image change was observed for more than 1-min exposure, preventing any change by the beam exposure during the measurement.

The angle-averaged spatial autocorrelation function \( \langle G(r) \rangle \) of the DF-TEM images is the statistical correlation of two points separated by distance \( r \) and was calculated following the method of Giraldo-Gallo et al. (26). The characteristic length scale \( L \) from \( \langle G(r) \rangle \) represents \( \xi \) in the disordered phase, although it approximately corresponds to the inverse square root of the paired dislocation density in the quasi-ordered crystalline phase. Each data point in Figs. 2 and 4 was calculated from the correlation functions of multiple (>10) images acquired from the membranes of the same thickness.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/3/11/eaao5173/DC1

fig. S1. Growth of SrTiO$_3$/Sr$_3$Al$_2$O$_6$ films.
fig. S2. Transfer of SrTiO$_3$ membranes.
fig. S3. Chemical stability of SrTiO$_3$ films.
fig. S4. Strain test using AFM force spectroscopy.
fig. S5. In situ heating experiment in TEM.
Control experiments
REFERENCES AND NOTES

1. N. D. Mermin, H. Wagner, Absence of ferromagnetism or antiferromagnetism in one- or two-dimensional isotropic Heisenberg models. Phys. Rev. Lett. 17, 1133–1136 (1966).

2. V. Berezinskii, Destruction of long-range order in one-dimensional and two-dimensional systems having a continuous symmetry group I. Classical systems. Sov. Phys. JETP 32, 493–500 (1971).

3. J. M. Kosterlitz, D. J. Thouless, Ordering, metastability and phase transitions in two-dimensional systems. J. Phys. C Solid State Phys. 6, 1181–1203 (1973).

4. Nobelprize.org, The Nobel Prize in Physics 2016 (2016); http://www.nobelprize.org/nobel_prizes/physics/laureates/2016/.

5. B. I. Halperin, D. R. Nelson, Theory of two-dimensional melting. Phys. Rev. Lett. 41, 121–124 (1978).

6. D. R. Nelson, B. I. Halperin, Dislocation-mediated melting in two dimensions. Phys. Rev. B 19, 2457–2484 (1979).

7. A. P. Young, Melting and the vector Coulomb gas in two dimensions. Phys. Rev. B 19, 1855–1866 (1979).

8. M. R. Beasley, J. E. Mooij, T. P. Orlando, Possibility of vortex-antivortex pair dissociation in two-dimensional superconductors. Phys. Rev. Lett. 42, 1165–1168 (1979).

9. K. J. Strandburg, Two-dimensional melting. Rev. Mod. Phys. 60, 161–207 (1988).

10. D. J. Bishop, J. D. Peppley, Study of the superfluid transition in two-dimensional \( ^3 \)He films. Phys. Rev. Lett. 40, 1727–1730 (1978).

11. D. J. Resnick, J. C. Garland, J. T. Boyd, S. Shoemaker, R. S. Newrock, Kosterlitz-Thouless transition in proximity-coupled superconducting arrays. Phys. Rev. Lett. 47, 1542–1545 (1981).

12. Z. Hadzibabic, P. Kröger, M. Cheneau, B. Battelier, J. Dalibard, Berezinskii–Kosterlitz–Thouless crossover in a trapped atomic gas. Nature 441, 1118–1121 (2006).

13. R. Reymen, S. Thiel, A. D. Cavigilia, L. F. Koukoutits, G. Hammerl, C. Richter, C. W. Schneider, T. Kopp, A. S. Rüetschi, D. Jaccard, M. Gabay, D. A. Muller, J.-M. Triscone, J. Mannhart, Superconducting interfaces between insulating oxides. Science 317, 1196–1199 (2007).

14. C. A. Murray, D. H. Van Winkle, Experimental observation of two-stage melting in a classical two-dimensional screened Coulomb system. Phys. Rev. Lett. 58, 1200–1203 (1987).

15. A. K. Geim, K. S. Novoselov, The rise of graphene. Nat. Mater. 6, 183–191 (2007).

16. J. C. Meyer, A. K. Geim, I. M. Katsnelson, K. S. Novoselov, T. J. Booth, S. Roth, The structure of suspended graphene sheets. Nature 446, 60–63 (2007).

17. A. Fasolino, J. H. Lux, M. I. Katsnelson, Intrinsic ripples in graphene. Nat. Mater. 6, 858–861 (2007).

18. J. Briñov, D. T. L. Alexander, A. Kis, Ripples and layers in ultrathin MoS\(_2\) membranes. Nano Lett. 11, 5148–5151 (2011).

19. N. Erdman, L. D. Marks, SrTiO\(_3\)(0 0 1) surface structures under oxidizing conditions. Surf. Sci. 526, 107–114 (2003).

20. M. Kawasaki, K. Takahashi, T. Maeda, R. Tsuchiya, M. Shinohara, O. Ishiyama, T. Yonezawa, M. Yoshimoto, H. Koimina, Atomic control of the SrTiO\(_3\) crystal surface. Science 266, 1540–1542 (1994).

21. D. Lu, D. J. Baek, S. S. Hong, L. F. Koukoutits, Y. Hikita, H. Y. Hwang, Synthesis of freestanding single-crystal perovskite films and heterostructures by etching of sacrificial water-soluble layers. Nat. Mater. 15, 1255–1260 (2016).

22. L. Pellegrino, M. Biasotti, E. Bellingeri, C. Bernini, A. S. Siri, D. Marré, All-oxide crystalline microelectomechanical systems: Bending the functionalities of transition-metal oxide thin films. Adv. Mater. 21 2377–2381 (2009).

23. P. Y. Huang, C. S. Ruiz-Vargas, A. M. van der Zande, W. S. Whitney, M. P. Levendov, J. W. Kevek, S. Garg, J. S. Alden, C. J. Hustedt, Y. Zhu, J. Park, P. L. McEuen, D. A. Muller, Grains and grain boundaries in single-layer graphene atomic patchwork quilts. Nature 469, 389–392 (2010).

24. C. Gerber, D. Anselmetti, J. G. Bednorz, J. Mannhart, D. G. Schlom, Screw dislocations in high-T\(_c\) films. Nature 350, 279–282 (1991).

25. K. Szt, W. Speier, R. Carius, U. Zastrow, W. Beyer, Localized metallic conductivity and self-healing during thermal reduction of SrTiO\(_3\). Phys. Rev. Lett. 88, 075508 (2002).

26. P. Giraldello-Gallo, Y. Zhang, C. Parra, H. C. Manoharan, M. R. Beasley, T. H. Geballe, M. J. Kramer, I. R. Fisher, Stripe-like nanoscale structural phase separation in superconducting BaPb\(_1\)\(_x\)BiO\(_3\). Nat. Commun. 6, 8231 (2015).

27. C.-C. Chen, A. B. Herhold, C. S. Johnson, A. P. Alivisatos, Size dependence of structural metastability in semiconductor nanocrystals. Science 276, 398–401 (1997).

28. G. A. Somorjai, Y. Li, Introduction to Surface Chemistry and Catalysis (John Wiley & Sons, 2010).

29. G. Wulfsberg, Principles of Descriptive Inorganic Chemistry (University Science Books, 1991).

30. J. P. Sethna, Statistical Mechanics: Entropy, Order Parameters, and Complexity (Oxford Univ. Press, 2006).

31. J. T. Ye, S. Inoue, K. Kobayashi, Y. Kasahara, H. T. Yuan, H. Shimotani, Y. Iwasa, Liquid-gated interface superconductivity on an atomically flat film. Nat. Mater. 9, 125–128 (2010).

32. M. Osada, T. Sasaki, Two-dimensional dielectric nanosheets: Novel nanoelectronics from nanocrystal building blocks. Adv. Mater. 24, 210–228 (2012).

33. J. N. Coleman, M. Lotya, A. O’Neill, S. D. Bergin, P. J. King, U. Khan, K. Young, A. Gaucher, S. De, R. J. Smith, I. V. Shvets, S. K. Arora, G. Stanton, H.-Y. Kim, K. Lee, G. T. Kim, G. S. Duesberg, T. Hallam, J. J. Boland, J. J. Wang, J. F. Donegan, J. C. Grunlan, G. Moriarty, A. Shmeliov, R. J. Nicholls, J. M. Perkins, E. M. Grieseson, K. Theuwissen, D. W. McComb, P. D. Nellist, V. Nicolesi, Two-dimensional nanosheets produced by liquid exfoliation of layered materials. Science 331, 568–571 (2011).

34. C. Lee, X. Wei, J. W. Kysar, J. Hone, Measurement of the elastic properties and intrinsic strength of monolayer graphene. Science 321, 385–388 (2008).

35. H. S. Seung, D. R. Nelson, Defects in flexible membranes with crystalline order. Phys. Rev. A 38, 1005–1018 (1988).

Acknowledgments: We acknowledge L. Radzihovsky for valuable discussions and P. Giraldello-Gallo for technical support of data analysis. Funding: This work was supported by the Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, under contract no. DE-AC02-76SF00515 (membrane synthesis and testing) and the Gordon and Betty Moore Foundation’s Emergent Phenomena in Quantum Systems Initiative through grant no. GBMF4415 (characterization of crystalline order). Part of this work was performed at the Stanford Nano Shared Facilities, supported by the NSF under award no. ECCS-1542152. Author contributions: S.S.H. and H.Y.H. designed the experiment. S.S.H. and D.L. synthesized and characterized the materials. J.H.Y., S.Y. and H.Y.H. performed the experiments. S.S.H. and D.L. designed the experiment. S.S.H. and H.Y.H. wrote the paper with input from all coauthors. Competing interests: The authors declare that they have no competing interests. Data and materials availability: All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. Additional data related to this paper may be requested from the authors.

Submitted 28 July 2017 Accepted 19 October 2017 Published 17 November 2017 10.1126/sciadv.aao5173

Citation: S. S. Hong, J. H. Yu, D. Lu, A. F. Marshall, Y. Hikita, Y. Cui, H. Y. Hwang, Two-dimensional limit of crystalline order in perovskite membrane films. Sci. Adv. 3, eaao5173 (2017).