**EXPERIMENTAL DETAILS.**

The x-ray measurements were performed using a custom PLD/x-ray diffraction system installed in the G3 hutch at the Cornell High Energy Synchrotron Source. Auxiliary Fig.1 illustrates the experimental geometry. The surface shown is an atomic force microscopy (AFM) image taken after the deposition performed in Fig. 1(a) of the manuscript where \( \theta = 11.3 \) ML. As suggested by the figure, steps on the sample resulting from the surface miscut are always aligned perpendicular to the incident beam. The x-ray beam produces scattering in the \( q_{||} \) direction that is directly related to the island correlations on the surface. For these experiments, a monochromatic (\( \Delta E/E=1\% \)) 10.0 keV x-ray beam with \( 8 \times 10^{13} \) photons/sec/mm\(^2\) was slit down to produce a 1.0 mm \( \times \) 0.5 mm beam at the sample. The specular component of the reflected signal was attenuated using a tin absorber to prevent detector saturation. To optimize the signal-to-background ratio of the diffuse scattering, the experiment was performed near the \( L = 0.275 \) r.l.u. position on the crystal truncation rod. The in-plane diffuse surface scattering was monitored using a CCD area detector operating as a linear detector in streak mode. The time resolution of the experiment is limited by both the readout time of the detector (\( \approx 78 \) ms) and by the incident x-ray flux (100 – 200 ms). For these growth conditions, we are able to collect 18 images between laser pulses.

The depositions were performed by laser ablating a single crystal SrTiO\(_3\) target using a 100 MW/cm\(^2\) KrF excimer laser (248 nm). The target is located 6 cm from the substrate. The area of the laser spot on the target was approximately 3.7 mm\(^2\) with a fluence of 1.9 J/cm\(^2\). This configuration deposited \( \approx 0.09 \) ML/pulse at a laser repetition rate of 0.2 Hz, with a \( 2 \times 10^{-4} \) Torr partial pressure.
of O$_2$. The substrate temperature was measured using an optical pyrometer ($\lambda = 4.8 - 5.3 \mu$m, emissivity=0.8). The substrate preparation procedure employed[1, 2] produced a TiO$_2$ terminated surface, and AFM confirmed the presence of single unit cell high steps separating large atomically flat terraces.

The diffuse scattering peaks are the direct result of single unit cell high islands, which was confirmed by ex-situ AFM. An AFM image after the deposition at 1000C, shown in Fig. 1(a) of the manuscript, is given in Auxiliary Fig. 2 confirming the presence of islands on the surface. The fast Fourier transform (FFT) of the image is shown in the inset. The presence of the diffuse Henzler ring is a direct result of the correlated islands on the surface. Our experimental detected x-ray intensity is a cut through these Henzler rings. Coarsening, during the post deposition cool-down ($\approx$ 3 hours), has caused the increase of the measured length scale of the AFM relative to the x-ray data.