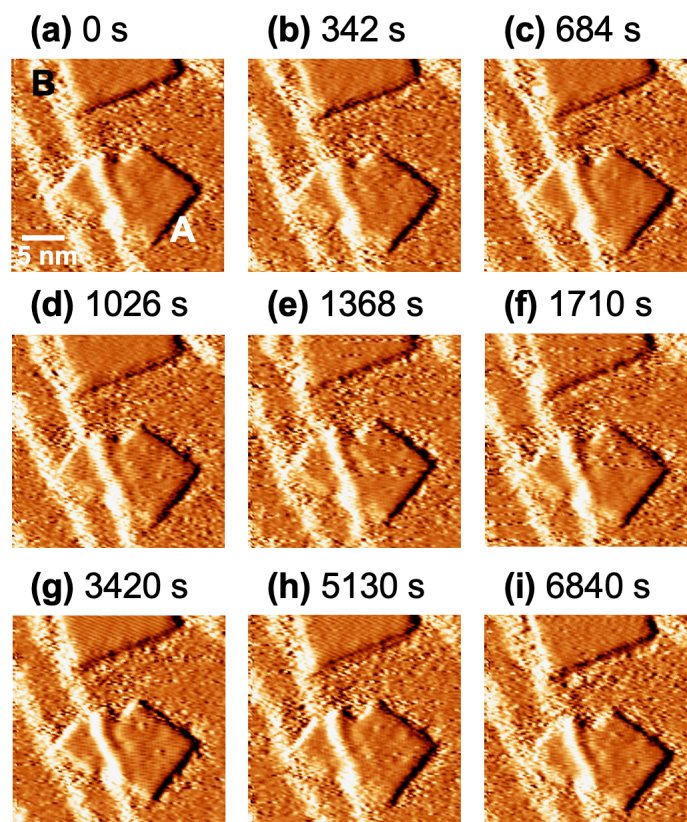# Supporting Information for

In situ Observation of Atomic Scale Growth of NaCl Thin Crystal on Au(111) by Scanning Tunneling Microscopy

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(a) 0 s **(b)** 342 s **(c)** 684 s

(d) 1026 s **(e)** 1368 s **(f)** 1710 s

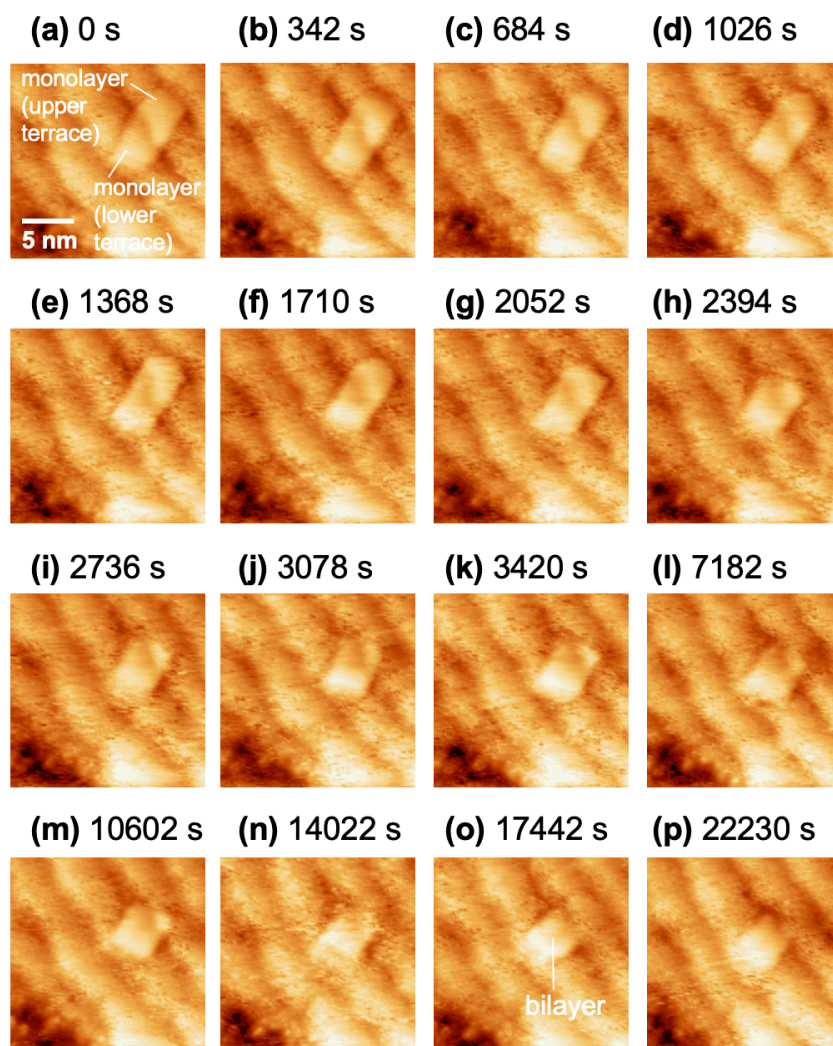**(g)** 3420 s **(h)** 5130 s **(i)** 6840 s

Figure S1 Sequential STM images showing the stability of the NaCl bilayers on Au(111) at room temperature. These images are the same as those in Figure 5, but displayed via the derivative processing. (a) An STM image focused on the NaCl bilayers indicated by A and B in Figure 4. (b)–(i) STM images obtained at the same area of (a) after the indicated time. All the STM images were obtained with V_s=0.5 V and I_t=0.5 nA.
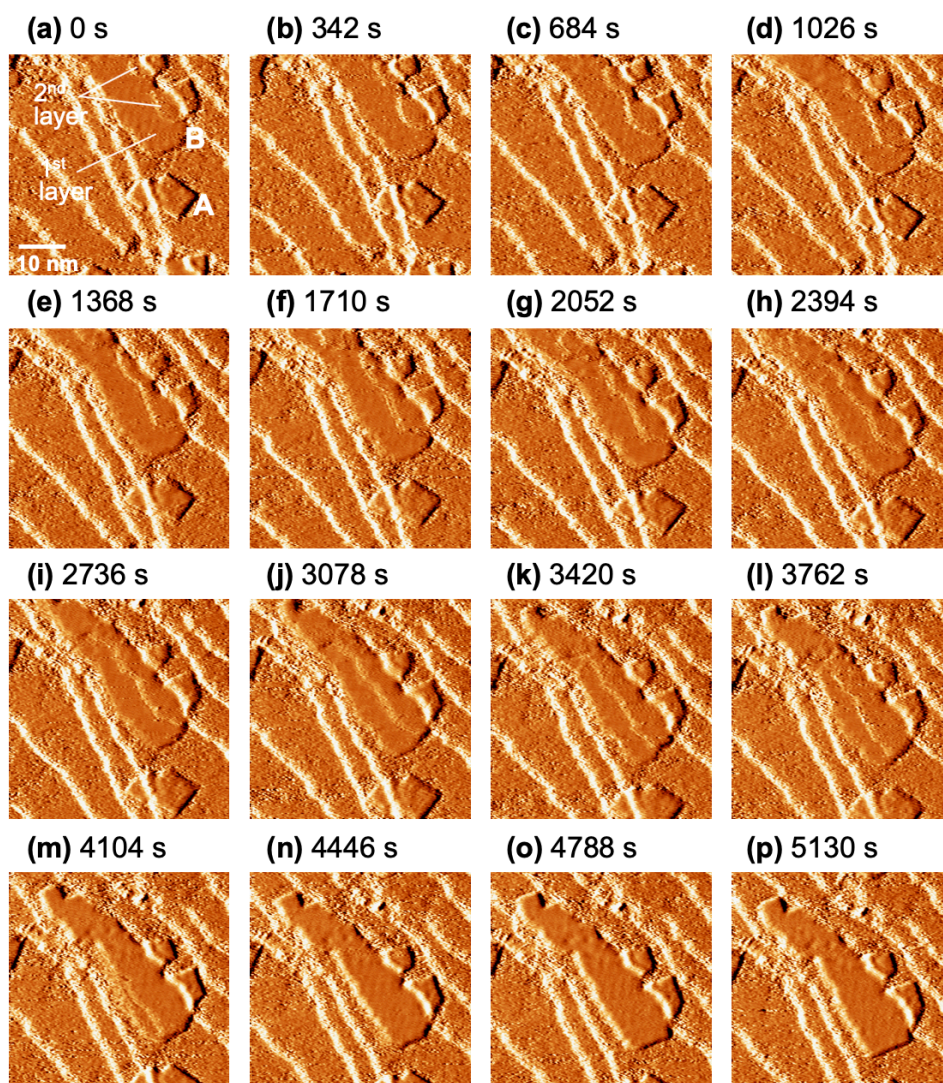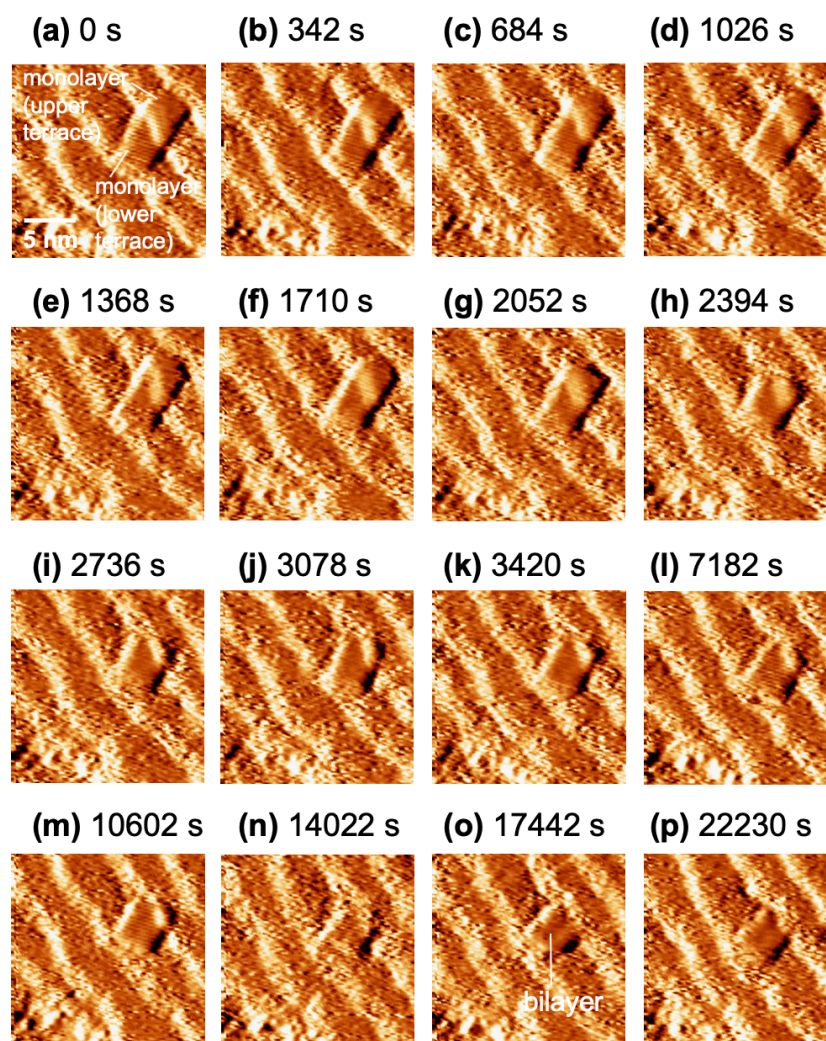
Figure S2 Sequential STM images showing the NaCl bilayer formation on the NaCl monolayer island on Au(111) at room temperature. These images are the same as those in Figure 6, but displayed via the derivative processing. (a) An STM image focused on the NaCl island indicated by B in Figure 4. For comparison, the NaCl bilayer indicated by A is also shown. (b)–(p) STM images obtained at the same area of (a) after the indicated time. All STM images were obtained with V_s=0.5 V and I_t=0.5 nA.

(a) 0 s   **(b)** 342 s   **(c)** 684 s   **(d)** 1026 s

monolayer (upper terrace)

monolayer (lower terrace)

5 nm

(e) 1368 s   **(f)** 1710 s   **(g)** 2052 s   **(h)** 2394 s

(i) 2736 s   **(j)** 3078 s   **(k)** 3420 s   **(l)** 7182 s

(m) 10602 s   **(n)** 14022 s   **(o)** 17442 s   **(p)** 22230 s
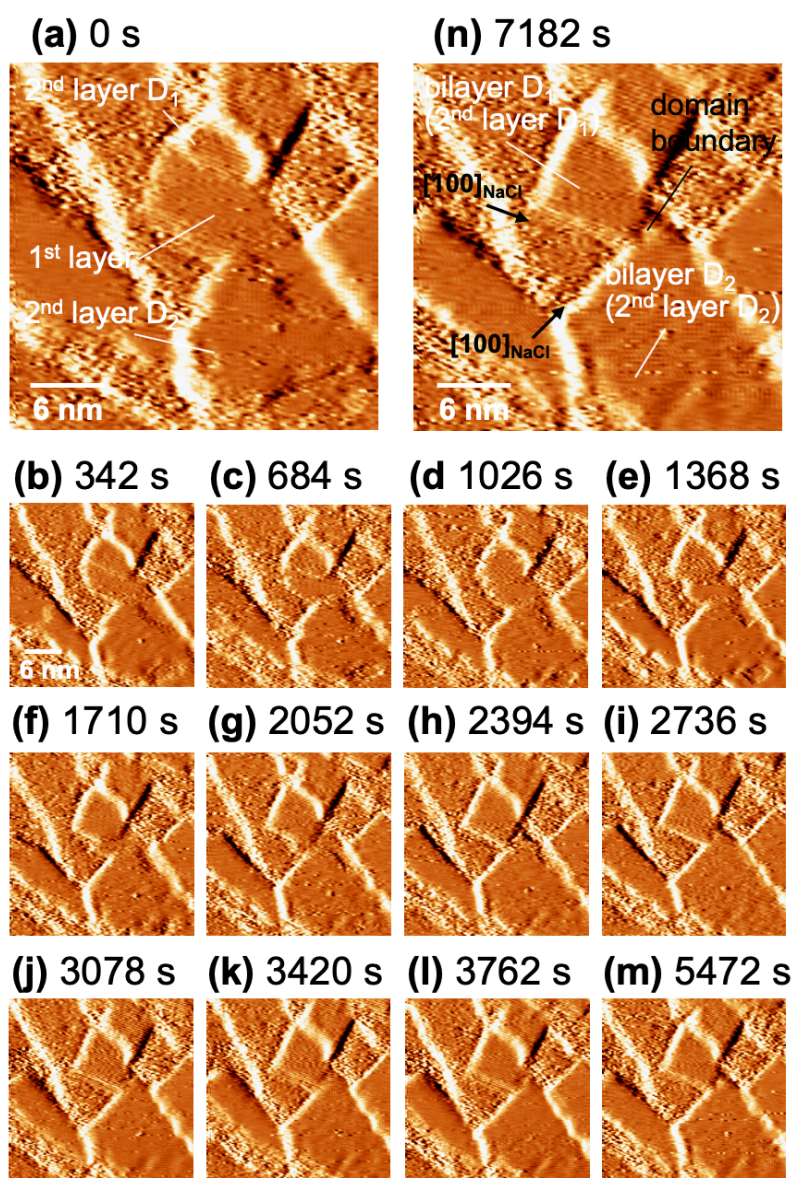
bilayer

Figure S3 Sequential STM images showing the NaCl bilayer formation on the NaCl monolayer island on Au(111) at room temperature. (a) An STM image focused on the NaCl island indicated by F in Figure 4. (b)-(p) The STM images obtained at the same area of (a) after the indicated time. The NaCl monolayer on the upper terrace was transferred to the NaCl island on the lower terrace forming the bilayer. All STM images were obtained by V_s=0.5 V and I_t=0.5 nA.
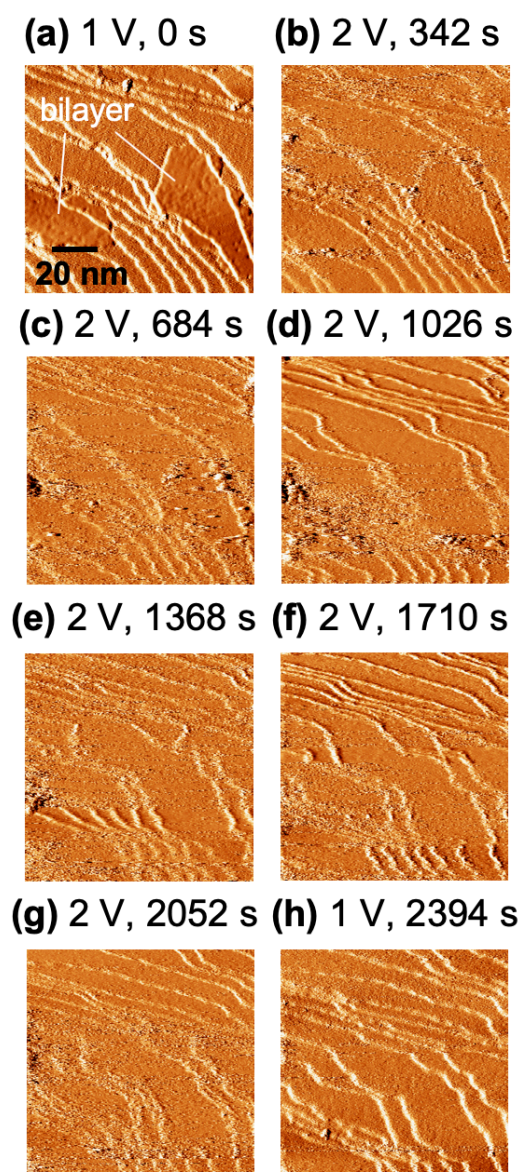
Figure S4   Sequential STM images showing the NaCl bilayer formation on the NaCl monolayer island on Au(111) at room temperature.   These images are the same as those in Figure S3, but displayed via the derivative processing.   (a) An STM image focused on the NaCl island indicated by F in Figure 4.   (b)-(p) The STM images obtained at the same area of (a) after the indicated time.   The NaCl monolayer on the upper terrace was transferred to the NaCl island on the lower terrace forming the bilayer. All STM images were obtained by V_s=0.5 V and I_t=0.5 nA.

(a) 0 s

(n) 7182 s

(b) 342 s   **(c)** 684 s   **(d** 1026 s   **(e)** 1368 s

(f) 1710 s   **(g)** 2052 s   **(h)** 2394 s   **(i)** 2736 s

(j) 3078 s   **(k)** 3420 s   **(l)** 3762 s   **(m)** 5472 s

Figure S5   Sequential STM images showing the formation of two NaCl bilayers on the NaCl monolayer island on Au(111) at room temperature.   These images are the same as those in Figure 8, but displayed via the derivative processing.   (a) An STM image focused on the NaCl island indicated by D in Figure 4.   (b)-(n) STM images obtained at the same area of (a) after the indicated time.   The NaCl bilayers, having different crystal orientations, formed on the same NaCl monolayer.   All STM images were obtained by V_s=0.5 V and I_t=0.5 nA.

(a) 1 V, 0 s   **(b)** 2 V, 342 s

bilayer

20 nm

(c) 2 V, 684 s   **(d)** 2 V, 1026 s

(e) 2 V, 1368 s   **(f)** 2 V, 1710 s

(g) 2 V, 2052 s   **(h)** 1 V, 2394 s

Figure S6 (a) An STM image of NaCl bilayers on Au(111) at room temperature. Scanning conditions: V_s=1.0 V, I_t=0.5 nA. These images are the same as those in Figure 9, but displayed via the derivative processing. (b)–(g) Sequential STM images obtained at the same area of (a) after the indicated time. The NaCl bilayers disappeared from the Au surface after the high-bias-voltage (2 V) scanning. Scanning conditions: V_s=2.0 V, I_t=0.5 nA. (h) An STM image obtained after (g) with low bias voltage. Scanning conditions: V_s=1.0 V, I_t=0.5 nA.

Movie 1   Sequential STM images of NaCl/Au(111) at room temperature.   All STM images were obtained by V_s=0.5 V and I_t=0.5 nA.   We measured STM images in a given period of time (342 s per frame).   Area: 100 nm×100 nm.   File format: MPEG4.

Movie 2   Sequential STM images of NaCl/Au(111) at room temperature.   This movie is the same as Movie 1, but displayed via the derivative processing.   STM images were obtained by V_s=0.5 V and I_t=0.5 nA.   We measured STM images in a given period of time (342 s per frame).   Area: 100 nm×100 nm. File format: MPEG4.