

**Supplemental Material for: Hydrogen-bonded one-dimensional  
molecular chains on ultrathin insulating films: Quinacridone on  
KCl/Cu(111)**

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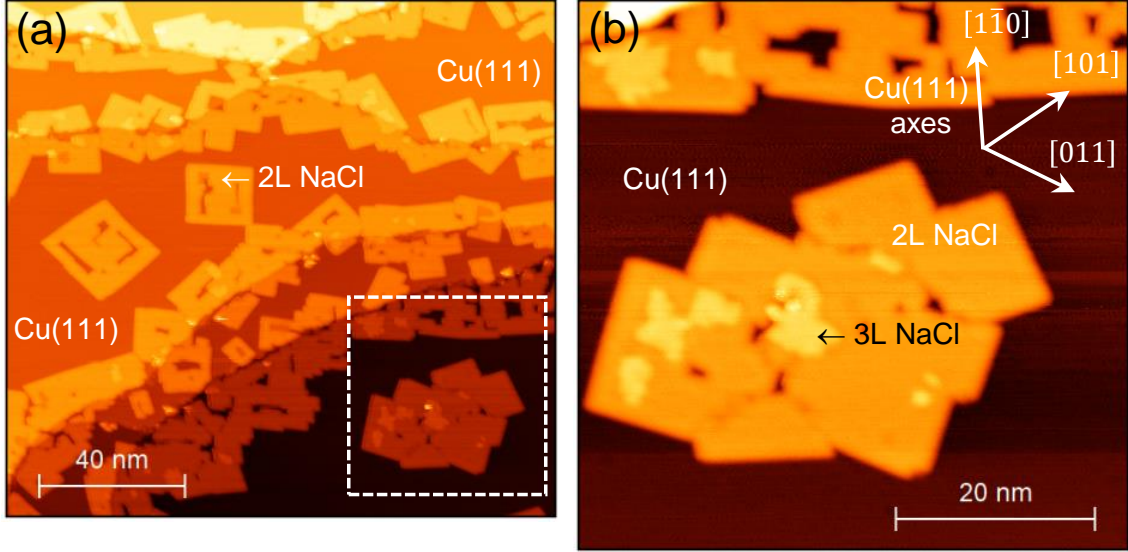


FIG. S1. STM topography images of an ultrathin NaCl film grown on Cu(111). STM parameters: sample bias 4.0 V, setpoint current 4 pA. (a,b) Two atomic layer (2L)-thick NaCl islands featuring a few three atomic layer (3L)-thick NaCl domains and surrounded by non-covered Cu(111) areas. (b) Same area as framed in dotted line in (a).

### A. Ultrathin NaCl films on Cu(111)

Figure S1 shows STM images of ultrathin sodium chloride (NaCl) films grown on Cu(111). NaCl is deposited for 5 minutes at a cell temperature of 833 K on a moderately cold sample about 5 minutes after the copper crystal is taken out of the low-temperature STM. The STM image shown in Fig. S1(b) has been used to measure the STM height of the 2L and 3L-thick NaCl areas relative to the Cu(111) surface and to compare it to the STM height of KCl islands grown in similar conditions on the same Cu(111) substrate.

### B. Two different relative orientations of KCl islands on Cu(111)

Figure S2 shows atomic-resolution STM images of the bare Cu(111) surface (in Fig. S2(a)) and of ultrathin (2L-thick) potassium chloride (KCl) films grown on Cu(111) (in Figs. S2(b) and S2(c)). For the KCl island shown in Fig. S2(b), one of the KCl  $\langle 110 \rangle$  directions is parallel to one of the Cu  $\langle 110 \rangle$  directions. The KCl island shown in Fig. S2(c) is oriented with the KCl  $\langle 110 \rangle$  directions at  $15^\circ$  from the Cu  $\langle 110 \rangle$  directions.

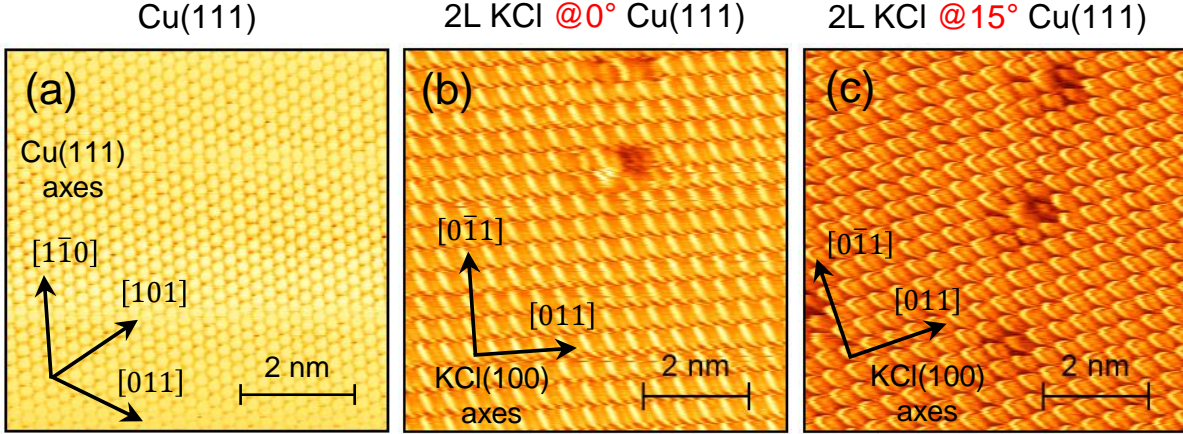


FIG. S2. STM topography images of (a) the bare Cu(111) surface and of (b,c) 2L-thick KCl islands on Cu(111) that have a different in-plane orientation from Cu(111) axes (see details in the text). STM parameters: (a) sample bias 0.3 V, setpoint current 1 nA; (b) 4.0 V, 30 pA; (c) 3.6 V, 10 pA. The STM image shown in (a) has been corrected for the lateral drift using the WSxM 4.0 software<sup>S1</sup>.

### C. QA chains on KCl islands rotated 15° from Cu(111) axes

Figure S3(a) shows an STM image of the sample, after deposition of QA molecules on an ultrathin KCl film grown on Cu(111). The ultrathin KCl film consists in 2L-thick KCl islands oriented with the KCl  $\langle 110 \rangle$  directions at 15° from the Cu  $\langle 110 \rangle$  directions. The adsorbed QA molecules have spontaneously assembled into chains on the bare Cu(111) surface as well as on the 2L-thick KCl islands. The density of adsorbed QA molecules is the highest on bare copper, presumably because the diffusion rate of QA on KCl is higher than on Cu(111).

Figures S3(b) and S3(c) show STM images of one of the 2L-thick KCl islands shown in Fig. S3(a), measured with a higher lateral resolution. The uncovered KCl areas exhibit a 2D moiré pattern, whose period is about 10.7 Å. The STM image shown in Fig. S3(c) reveals the presence of structural disorder in some of the QA chains. Nevertheless, two favored in-plane orientations clearly appear, which correspond to  $\langle 100 \rangle$  directions of KCl. In order to confirm this result and compare it to the orientation of the QA chains grown on bare copper, we measure in Fig. S3(a) the in-plane orientation of about 300 molecular chains on bare copper and 2L KCl/Cu(111). Figures S3(d) and S3(e) show the results in two polar histograms, where the chain orientation is defined as the angle between the chain axis and

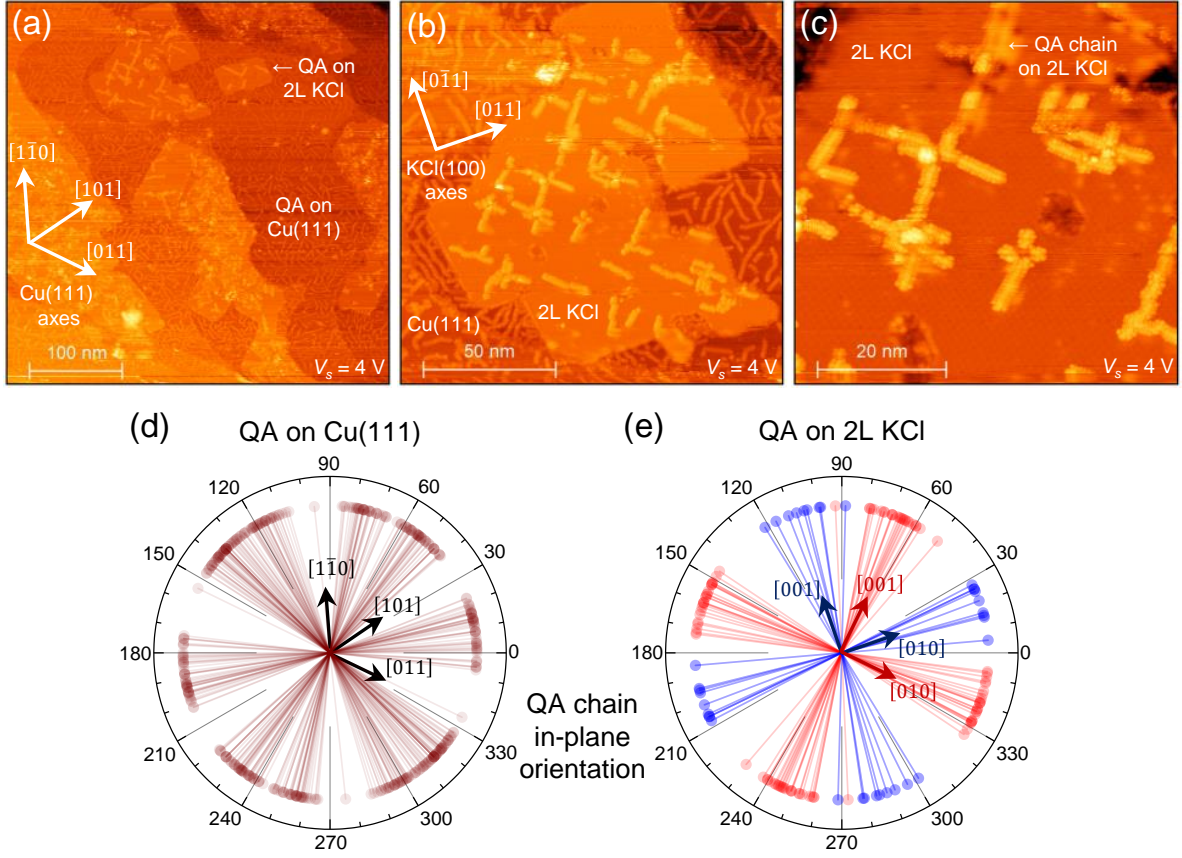


FIG. S3. (a-c) STM topography images of QA molecules on an ultrathin KCl film grown on Cu(111), measured at a sample bias  $V_s = 4.0$  V and a setpoint current (a,b)  $I_t = 50$  pA or (c)  $I_t = 20$  pA. (d,e) Polar histograms of the QA chain in-plane orientation (d) on bare Cu(111) and (e) on 2L-thick KCl/Cu(111), obtained from the analysis of the STM image shown in (a). In (e), different colors (red and blue) are used to distinguish 2L-thick KCl islands that have two different in-plane orientations with respect to the underlying Cu(111) surface.

the fast-scanning axis of the STM (i.e., the horizontal in Fig. S3). The graph in Fig. S3(d) reveals that QA chains on bare copper are oriented along six preferential directions, which are at  $\pm 20^\circ$  from the main crystal axes of the Cu(111) surface. This result is in agreement with previously reported STM observations of QA molecular arrangements on Cu(111) after deposition at room temperature in the submonolayer coverage range<sup>S2</sup>.

In Fig. S3(e), we consider two distinct 2L-thick KCl islands that do not have the same in-plane orientation with respect to the Cu(111) surface. We use a different color for the QA chains grown on each of these two KCl islands. Thus, we demonstrate that the QA chains

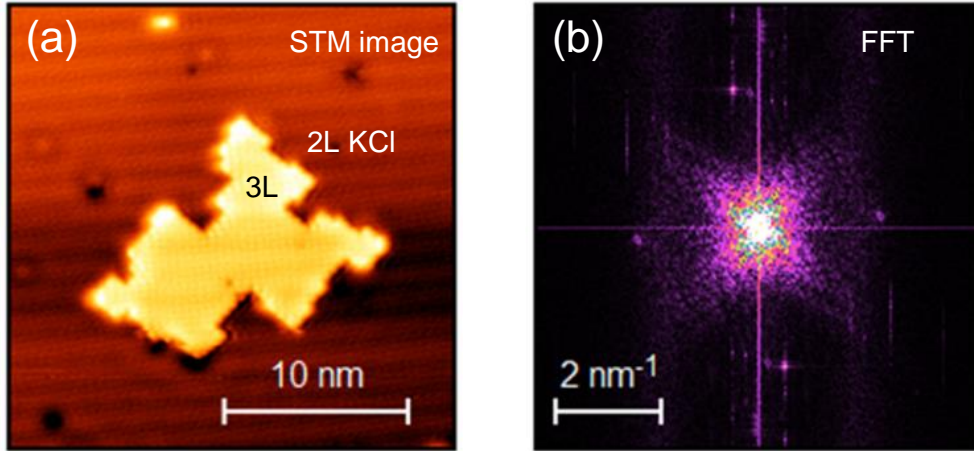


FIG. S4. (a) STM topography image of an ultrathin KCl film grown on Cu(111) featuring two (2L) and three (3L) atomic layer-thick areas. STM parameters: 3.0 V, 4 pA. The moiré pattern and the atomic resolution in the 3L-thick KCl area are visible. (b) Fast Fourier transform (FFT) of the STM image shown in (a).

on KCl are oriented along two favored directions, which are parallel to  $\langle 100 \rangle$  directions of the corresponding KCl island. This shows that, on these 2L-thick KCl islands, the in-plane orientation of the QA chains is determined by interactions between the adsorbed molecules and the KCl(100) surface, and not by the underlying Cu(111) surface. These observations also confirm that the same favored in-plane orientations of the QA chain axis relative to the KCl lattice is observed, irrespective of the in-plane orientation of the KCl islands relative to the Cu(111) surface.

#### D. Atomic resolution in Fig. 2(e)

Figure S4(a) shows a portion of the STM image shown in Fig. 2(e), which corresponds to the same area as highlighted in the insert in Fig. 2(e). Figure S4(b) shows the fast Fourier transform (FFT) of the STM image shown in Fig. S4(a). The four bright and sharp spots visible in Fig. S4(b) indicate a four-fold rotational symmetry and a spatial period of about 4.4 Å, which corresponds to the quadratic periodic lattice of the (100) KCl surface, considering that only one type of ions (presumably the Cl<sup>-</sup> anions) of the KCl surface is imaged by the STM.

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