## **Supporting Information**

## Atomic-Scale Processes at Fluorite/Water Interface Visualized by Frequency Modulation Atomic Force Microscopy

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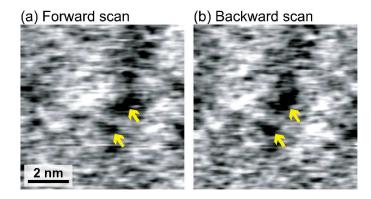
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## **Supporting Figures**



**Figure S1**: (a) Forward and (b) backward scan FM-AFM images of the atomically-disordered structures obtained in acidified saturated solution. The both images show similar atomic-scale contrasts as indicated by the arrows. The results suggest that the tip scan does not significantly disturb the atomic-scale surface structures.

## Supplementary Video Files

Following video files consist of successive FM-AFM images of fluorite(111) surface obtained in each solution condition.

- Water.avi: Pure water (pH = 6.5,  $500 \text{ nm} \times 500 \text{ nm}$ ).
- AcidifiedWater.avi: Acidified water (pH = 2, 1  $\mu$ m × 1  $\mu$ m).
- Saturated.avi: Saturated solution (pH = 6, 500 nm × 500 nm).
- AcidifiedSaturated.avi: Acidified saturated solution (pH = 2, 500 nm × 500 nm).
- Supersaturated 10. avi: Supersaturated solution with  $\sigma = 10$  (pH = 6, 500 nm × 500 nm).
- Supersaturated 100.avi: Supersaturated solution with  $\sigma = 100$  (pH = 6, 500 nm × 500 nm).

Note that  $\sigma$  denotes the degree of supersaturation. The time indicated in each frame corresponds to the time passed since the scan start. The time required for making the tip approach after the deposition of the imaging solution on the cleaved surface is typically 10 min.