

# Supplementary Materials: Growth of Calcite in Confinement

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## 1. Variation in growth at nominally equal conditions

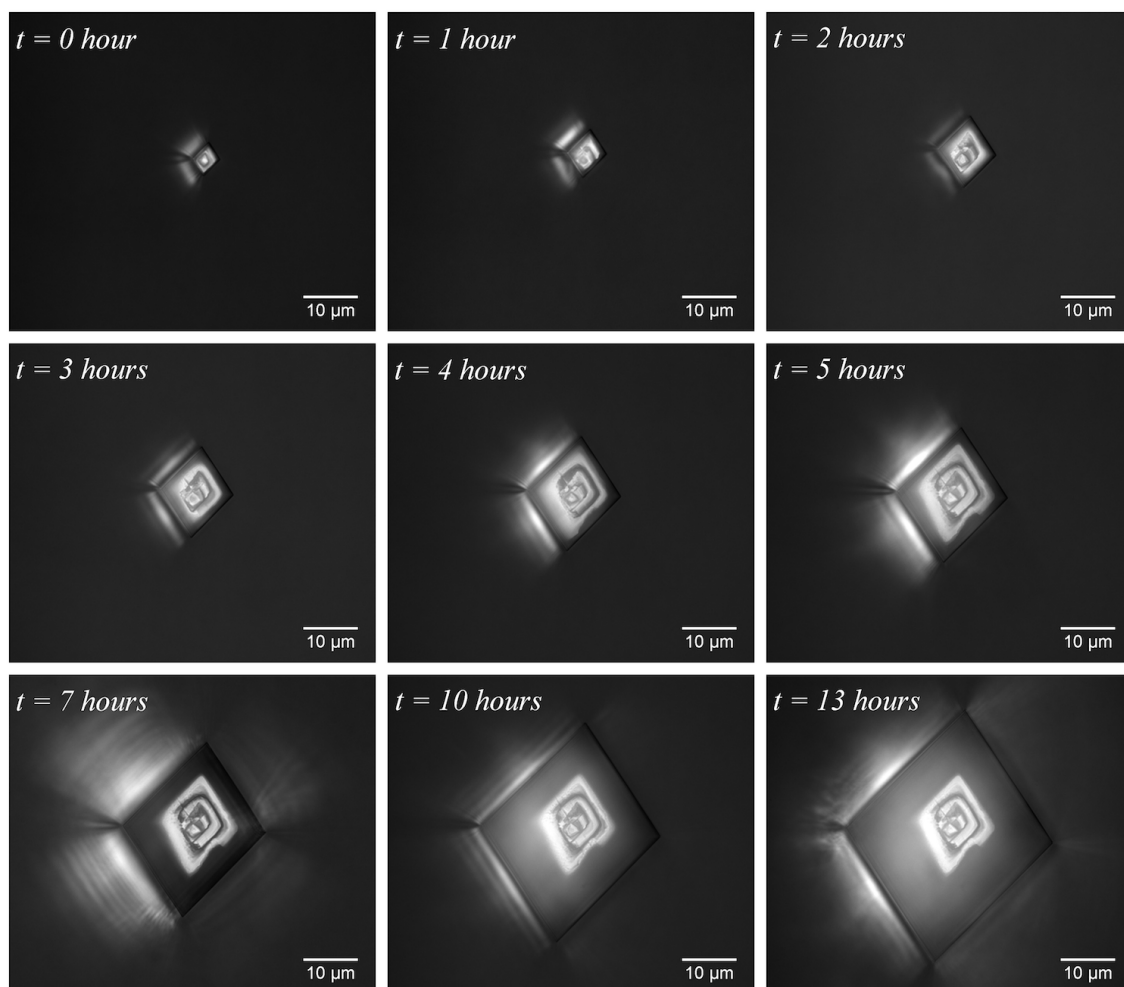
Very many (20-30) crystals have been grown in the microfluidic device presented in this paper. Crystal A and B represent the two main types of confined crystal growth found in this study: Smooth rim contacts that grow more slowly upwards and at some point in time stop growing upwards and rough rim contacts where different parts of the crystal grow intermittently fast and slow, but where the whole crystal is lifted up at a higher rate. In this section of the supplementary material we present images of the crystals for which we have quantified the growth rates and presented them in the main paper. We have also made available movies of the two crystals A and B.

For crystal A during the first 10 hours the vertical growth rate was 25 nm/h, the last 15 hours it was only 1.5 nm/h. The outwards growth rate was stable at  $310 \pm 9$  nm/h outwards during the entire 26 hours. It is a challenge to explain why the vertical growth rate changes even though the supersaturation and the outwards growth rate are constant.

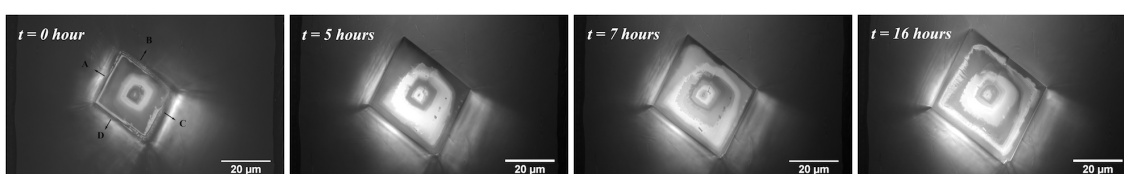
Crystals A and B may be considered representatives of two different families of behaviour that we have observed in 20-30 crystal growth experiments: smooth rim growth and intermittent rim growth, both rims actively lifting the crystal. These differences are not unique to calcite: similar topographies have been documented for confined growth rims of  $\text{NaClO}_3$  [1].

Crystal B grows almost 3 times faster outwards and 7 to 120 times faster upwards than crystal A even though the concentration  $c$  is 0.7 mM for B and 0.8 mM for A. Both crystals keep the overall rhombohedral calcite shape, but crystal B has several steps and other visible "defects" on the confined surface and on the lower right edge.

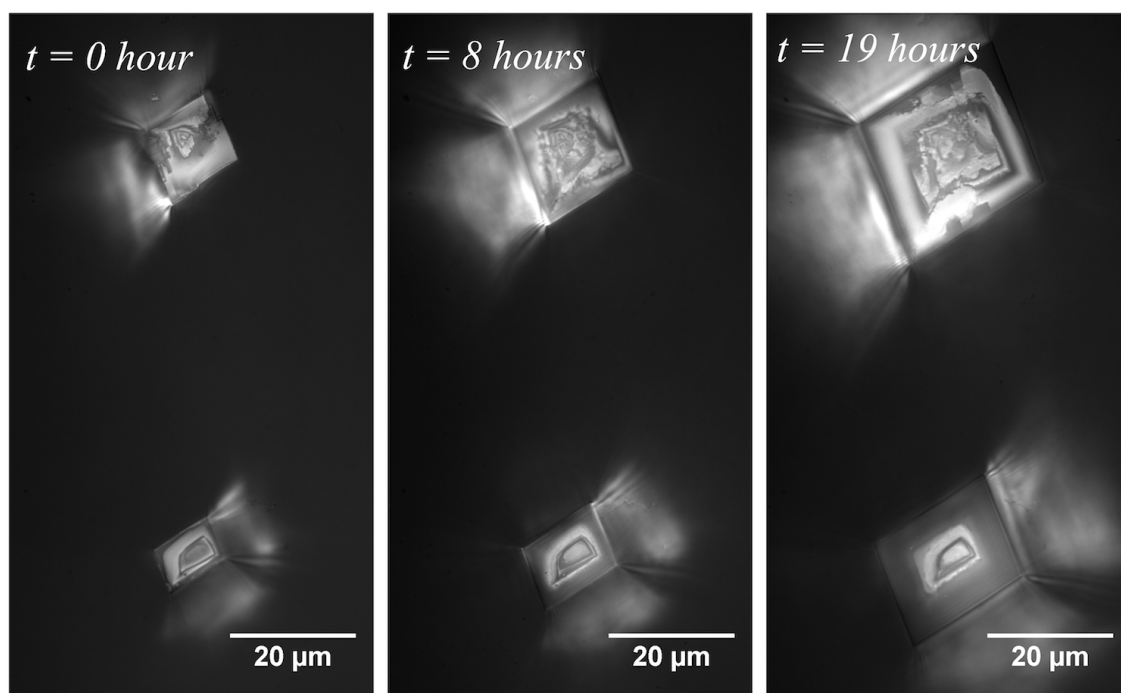
Crystal C in Figure S1 grows upwards at the rapid rate of 151 nm/h for the first three hours and then stops growing upwards almost completely. The inner shape and intensity of the confined surface remains constant and while the outer edge and rim width grows steadily. One may observe a very even, dark intensity around the whole contact rim signifying an even, close contact (small  $h$ ).



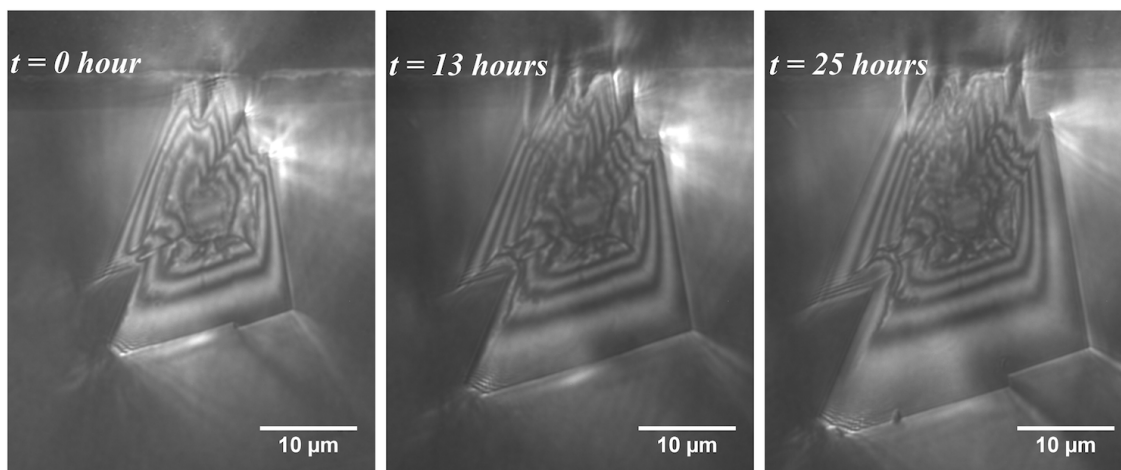
**Figure S1.** 13 hours growth of crystal C. The  $\text{Ca}^{2+}$  concentration here was 0.8 mM. Flow direction from bottom to top.



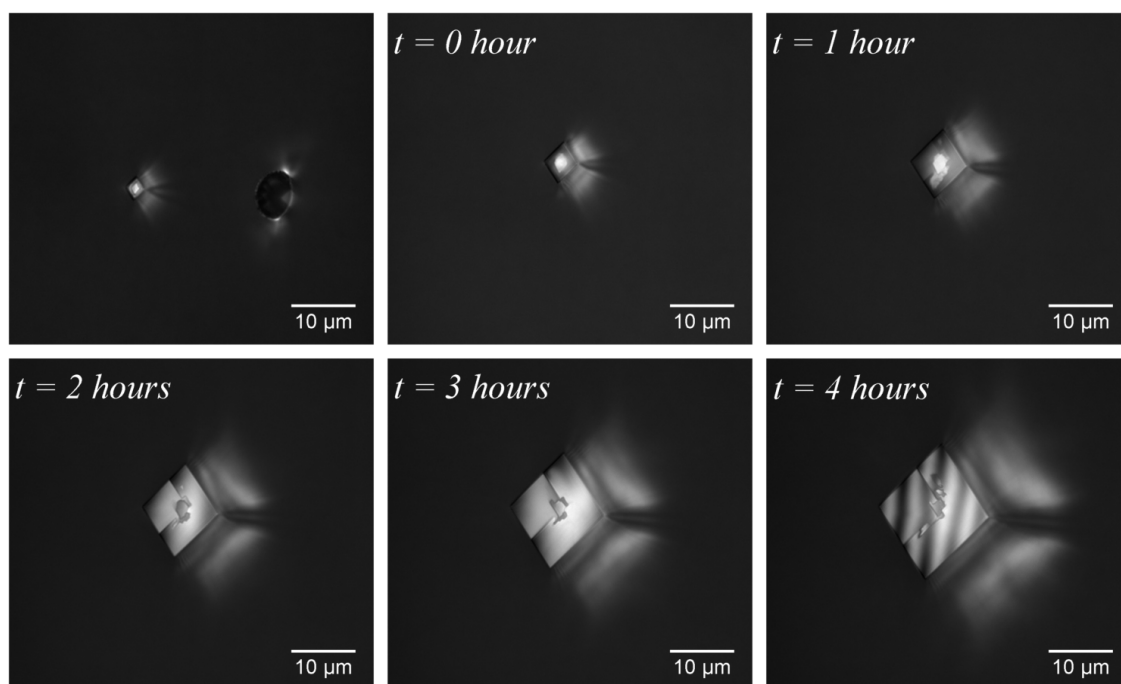
**Figure S2.** 16 hours growth of crystal D. The  $\text{Ca}^{2+}$  concentration here was 0.7 mM. Flow direction from bottom to top.



**Figure S3.** 19 hours growth of crystals E and F. The  $\text{Ca}^{2+}$  concentration here was 0.7 mM. This experiment shows the two classes of confined crystals contacts at the same time: intermittent (top) and smooth (bottom). Flow direction from bottom to top.



**Figure S4.** 25 hours growth of crystal G. The  $\text{Ca}^{2+}$  concentration here was 0.7 mM. Flow direction from left to right. The confined contact surface is not a  $10\bar{1}4$  surface



**Figure S5.** 4 hours growth of crystal H. The  $\text{Ca}^{2+}$  concentration here was 0.7 mM. The crystal seems initially smooth and flat. After 1 hour it develops a "defect" and continues to grow two separate flat surfaces at different heights and the crystal surface tilts with respect to the glass surface. Flow direction from bottom to top.

1. Røyne, A.; Dysthe, D.K. Rim formation on crystal faces growing in confinement. *Journal of Crystal Growth* **2012**, *346*, 89–100.