Rubrene on mica: from the early growth stage to late crystallization

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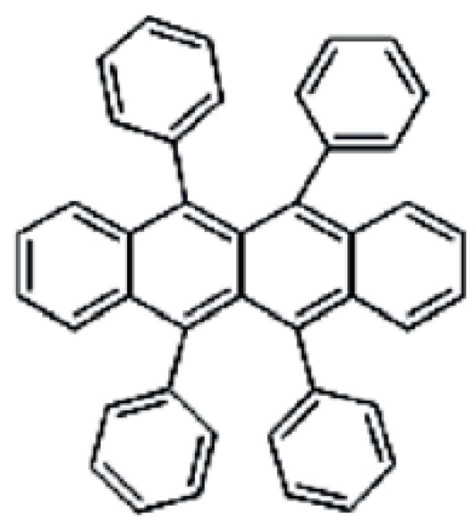
Motivation

Rubrene shows a high hole mobility in single crystalins. However, Rubrene thin films exhibit a much lower mobility. Recent work on Rubrene thin films on weakly interacting amorphous SiO_2 showed that the formed crystalline structures can increase the thin film mobility drastically [1,2].

Here, we present a detailed atomic-force microscopy (AFM) study on Rubrene thin films on the stronger interacting crystalline mica(001) surface. The evolution of the initial growth stage and pecularities in the growth rate are analyzed. For these intial layers of amorphous Rubrene the wetting behaviour is investigated as a function of deposition temperature. Finally, the morphology of the crystalline spherulites, developing in thick films is shown.

Experimental

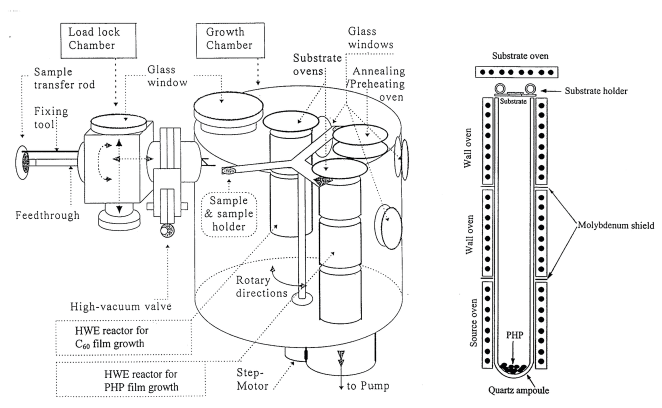
Rubrene



- Tetracene core
- p-type semiconductor
- single crystal mobility of 15 cm²/Vs
- difficult to grow thin films
  - very low mobility 10^{-6} cm²/Vs
- Amorphous phase not stable against O_2
- High stability of crystalline phase

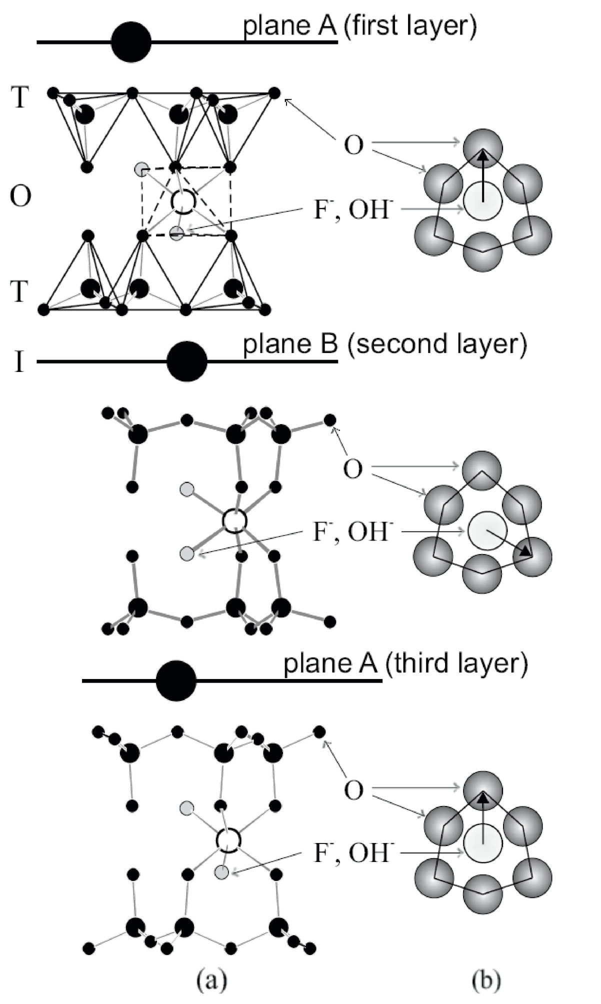
Hot Wall Epitaxy

Deposition temperature:
363 K to 393 K
Base pressure: $1x10^{-6}$ mbar
Source temperature:
453 K to 508 K
Wall temperature:
453 K to 508 K
Deposition rate: roughly 25nm/h

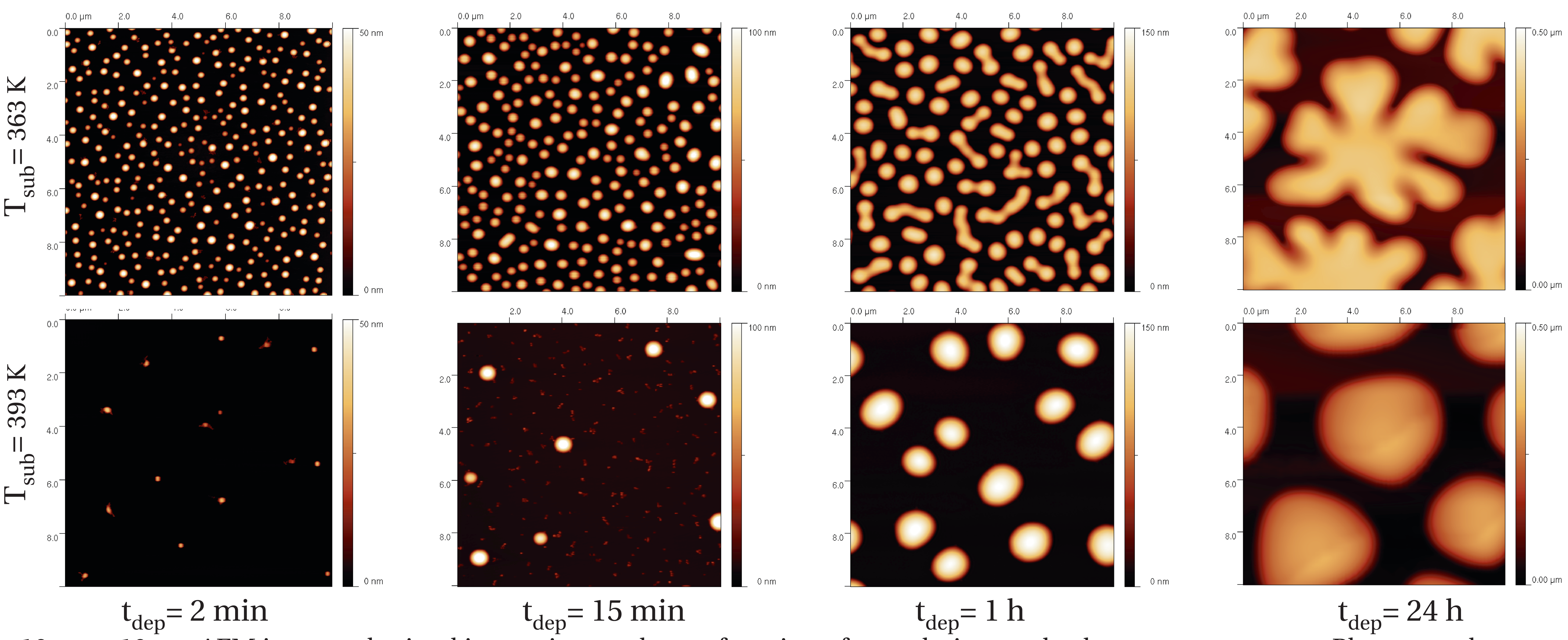


Mica(001)

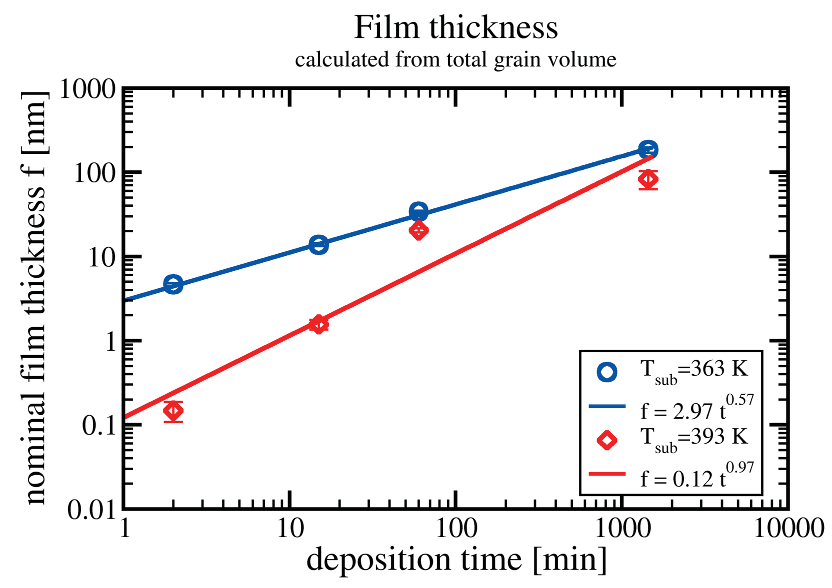
The mica(001) surface used is a cleavage plane of $2M_1$-muscovite with the formula $KAl_2(AlSi_3)O_{10}(OH,F)_2$. Due to charge repulsion between the oxygen in the top most layer the anions are displaced from the center of the hexagonal opening - which leads to a two fold symmetry of mica(001) and a strong surface dipol .

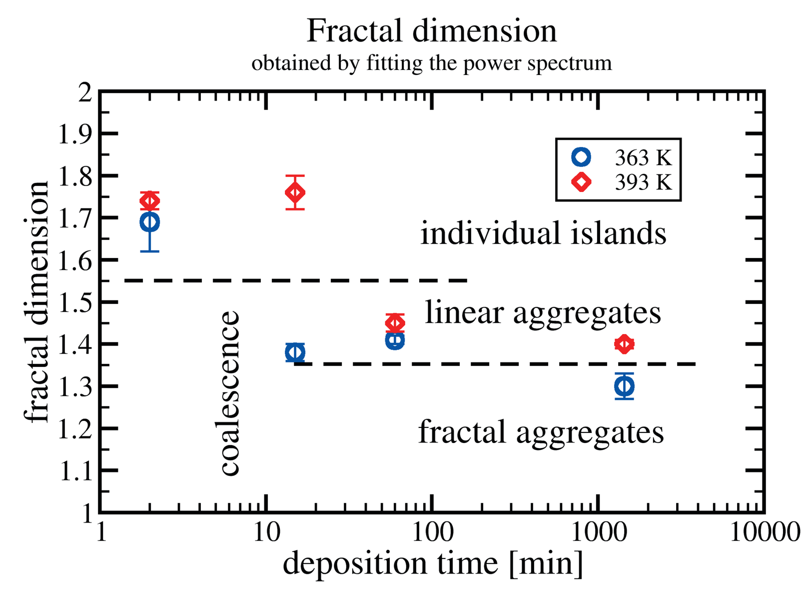


Morphological analysis



10 µm x 10 µm AFM images obtained in tapping mode as a function of growth time and substrate temperature. Please not the different height scales. Morphological evolution: Circular islands -> coalescence -> larger fractal islands

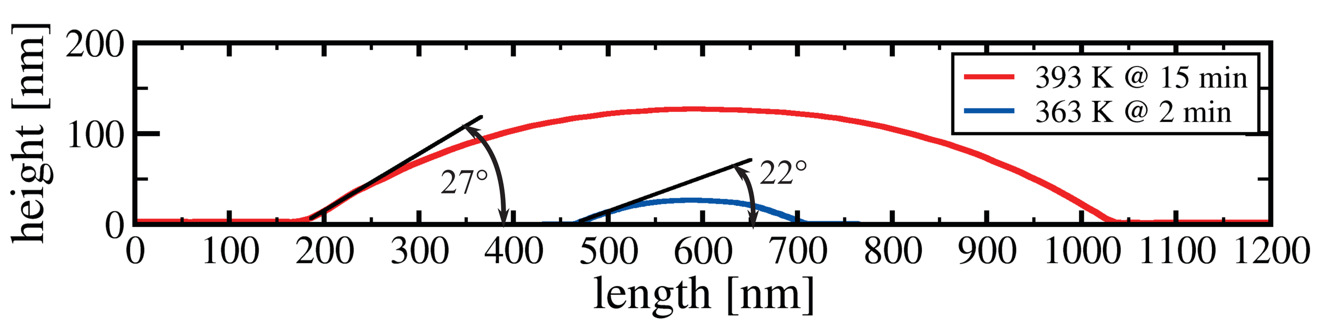




- Higher coverage at 363 K
  - P_{stick}(363 K) > P_{stick}(393 K)
- Sublinear growth rate at 363 K
  - P_{stick}(Rub./Rub.) < P_{stick}(Rub./mica)
- Fractal dimension D calculated from power spectra [3]
- Fractal analysis allows seperation into three stages
  - growth of individual islands: D > 1.5
  - begin of coalescence: 1.35 > D > 1.5
  - formation of branched islands: D < 1.35

Contact angle of Rubrene on mica(001)

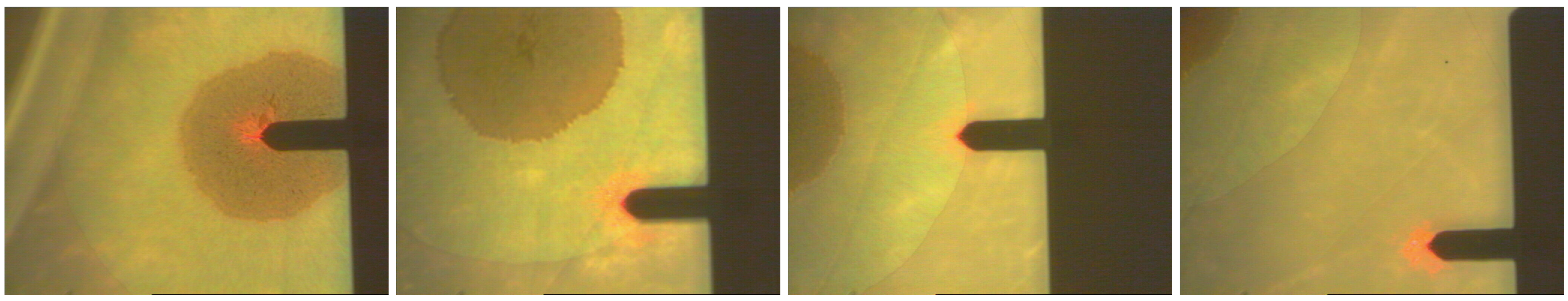
AFM Cross section (true aspect ratio)

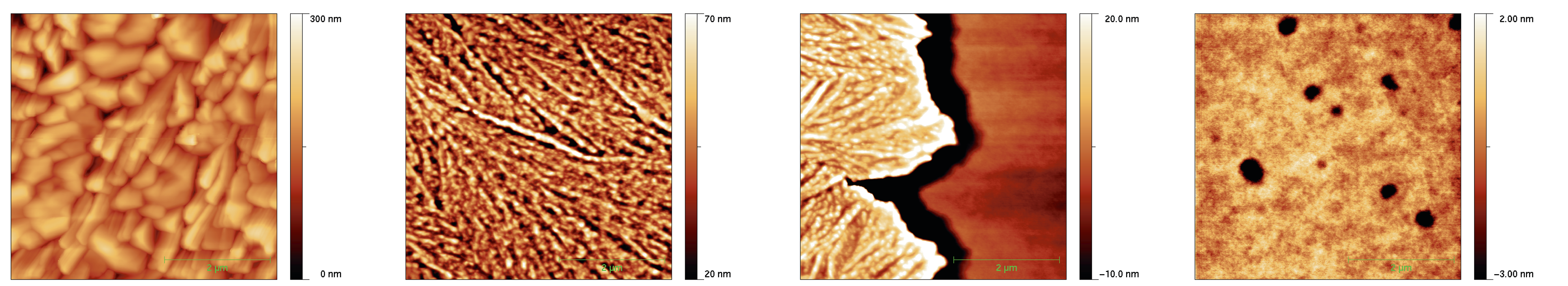


T_{sub}	Θ
363 K	22°
393 K	27°

- With increasing T_{sub} the contact angle Θ increases
  - System gets more rubrenophobic
  - Surf. energy of Rubrene increases stronger with increasing T_{sub} then surf. energy of mica.

Formation of crystalline spherulites





- Optical microscopy images (top row) of thicker films show crystalline spherulites embedded in a transparent amorphous matrix.
- T_{dep}=90°C, T_{source}=235°C (corresponds to a higher growth rate)

- Center: facetted large crystals showing a slight radial structure
- Iris: typical branched dentritic growth, strong radial structure
- Edge: deep gap between crystaline spherulite and matrix
- Outer region: amorphous film with some holes a few nm deep

Summary

Initial growth

- Amorphous islands start to coalesce with increasing coverage.
- The sticking coefficient depends on temperature and is different on mica and on Rubrene.
- Wettability of Rubrene on mica(001) decreases with increasing T_{sub}.

Later growth stage

- Formation of crystalline spherulites in an amorphous matrix.
- The spherulite center is formed by large facetted crystallites.
- The iris is characterized by branched dentritic crystallites with a strong radial structure.

References

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