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Real Time Diffuse and Specular Scattering for 3d Characterisation of Thin Film Growth

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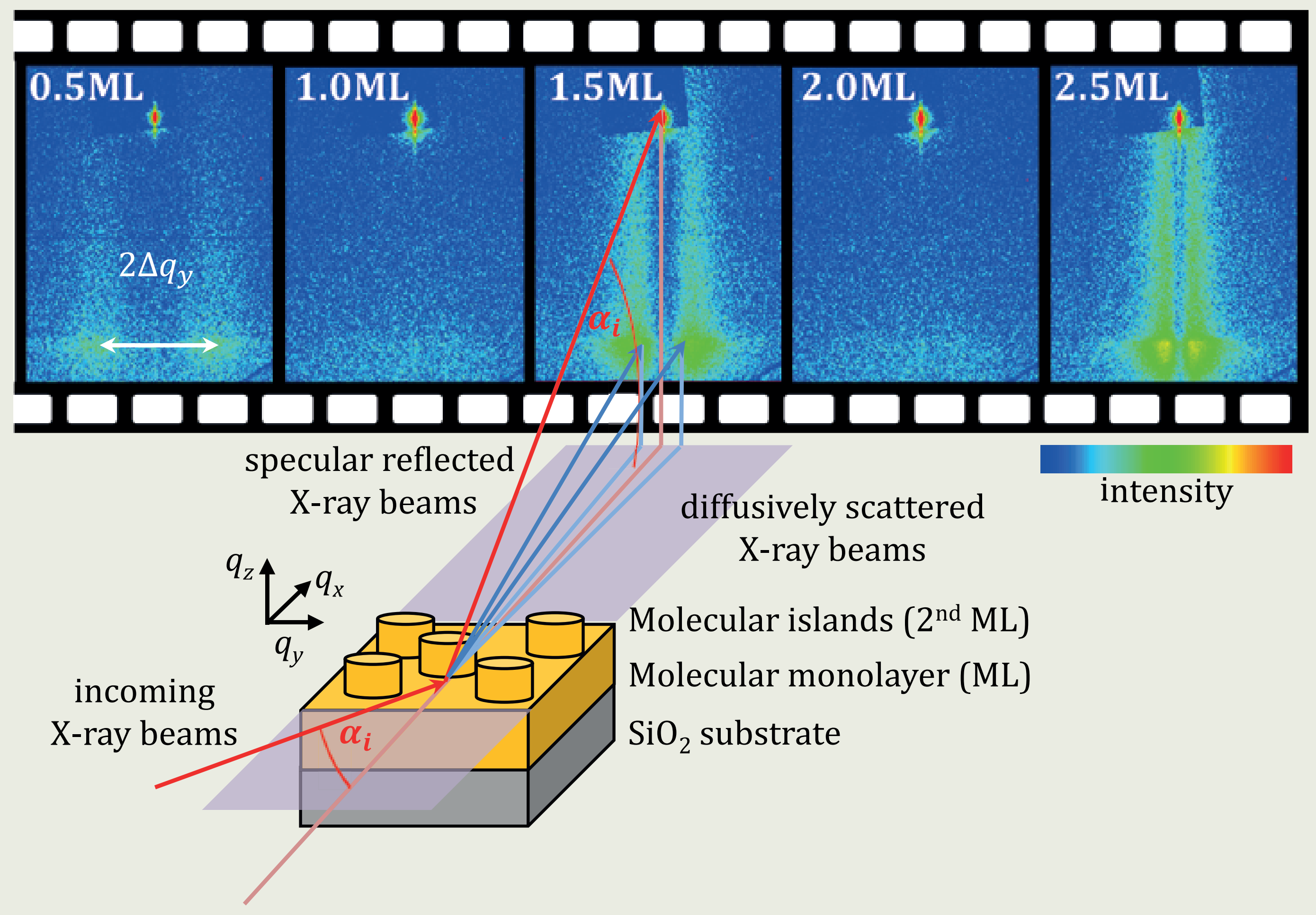
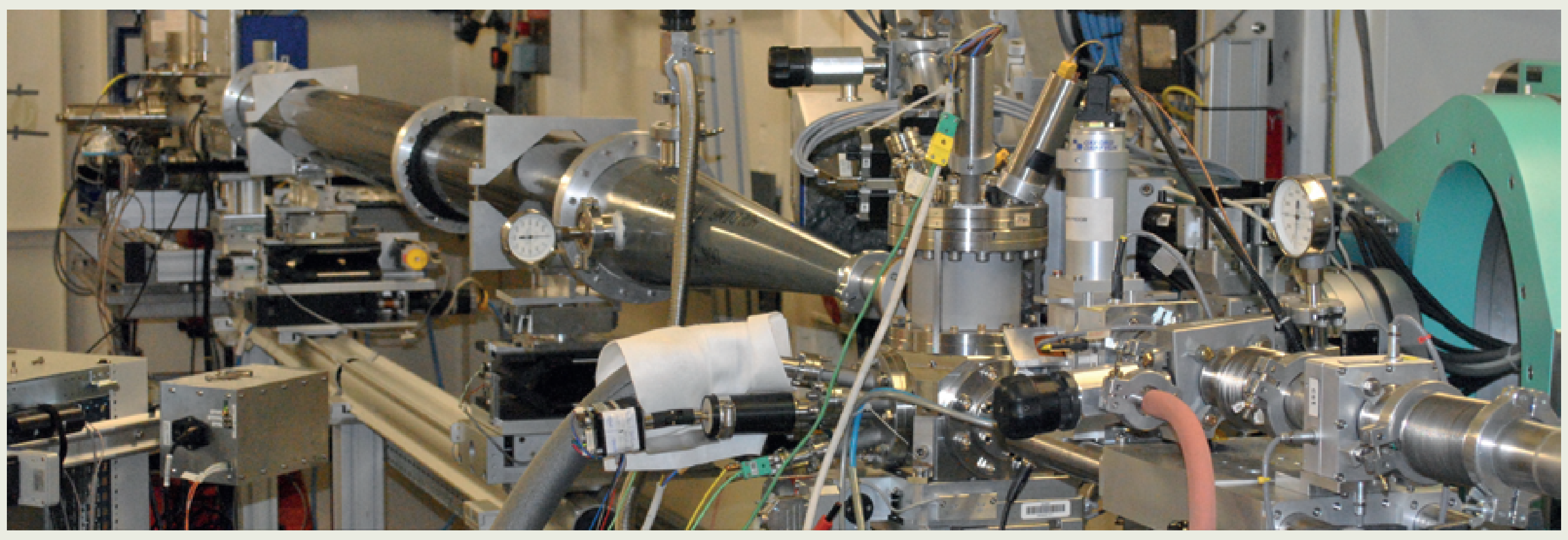
Scattering Geometry & Real-Time Data



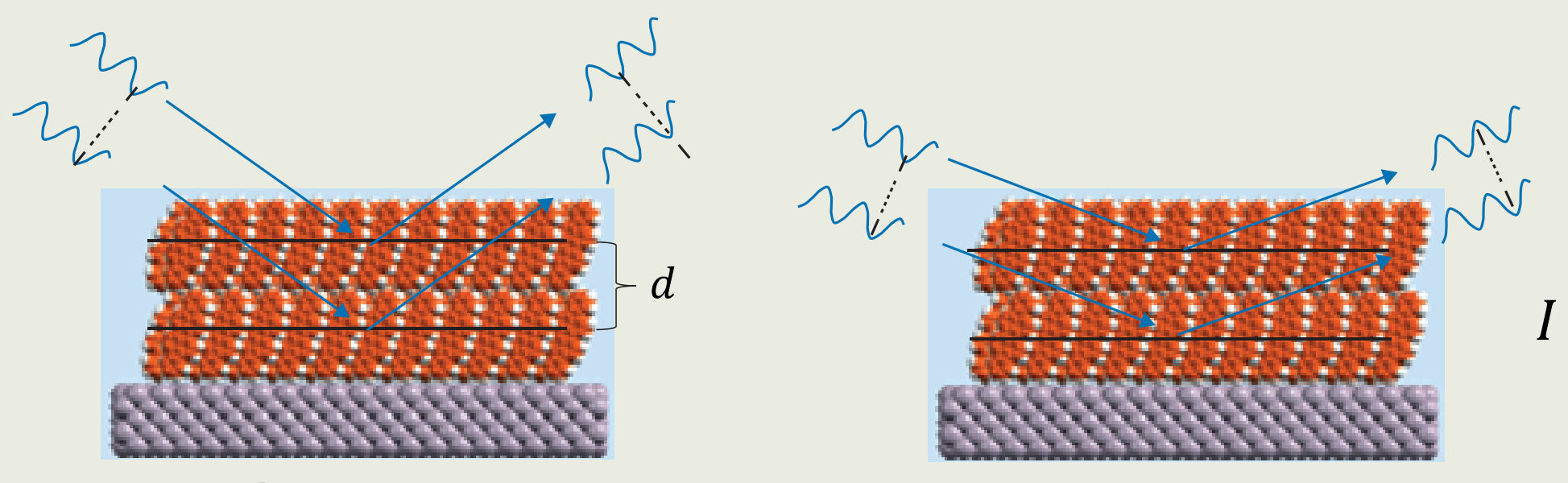
Simultaneous acquisition of growth oscillations and diffuse scattering ('GIXSAXS like') allows for a detailed description of the morphology evolution during organic thin film growth with high temporal resolution enabling to monitor and control fundamental growth kinetics on a submonolayer basis. [1] [2][3]

At ESRF the surface diffraction beamlines ID3 and ID10 are especially suited for this type of measurements. With its 4m evacuated flight path ID10 is equipped to resolve up to 1000nm large island structures.

For thin film growth we use a portable UHV chamber with beryllium window that can be mounted on the ID3 and ID10 diffractometers.



Specular: Evolution of Layer Coverages

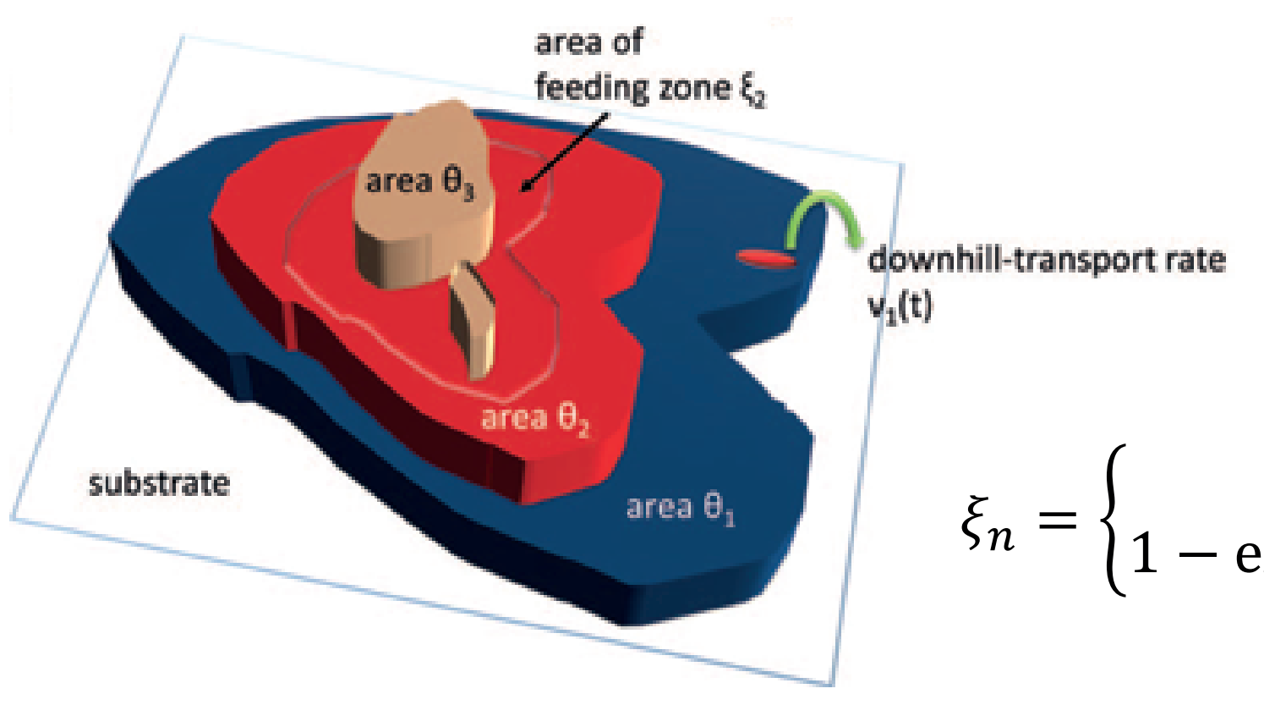


$$I = \left| f_{substrate} \cdot e^{-i\varphi} + f_{monolayer} \sum_n \theta_n(t) \cdot e^{-iq_z dn} \right|^2$$

θ_n: *Layer Coverage*

Monitoring X-ray growth oscillations, i.e. temporal oscillations of the X-ray reflectivity during thin film growth, is an important technique for *in situ* and real-time characterization of thin film growth. [4]

Analytical models may be applied to fit the oscillation to evaluate the monolayer coverages and the surface roughness evolution during thin film growth.



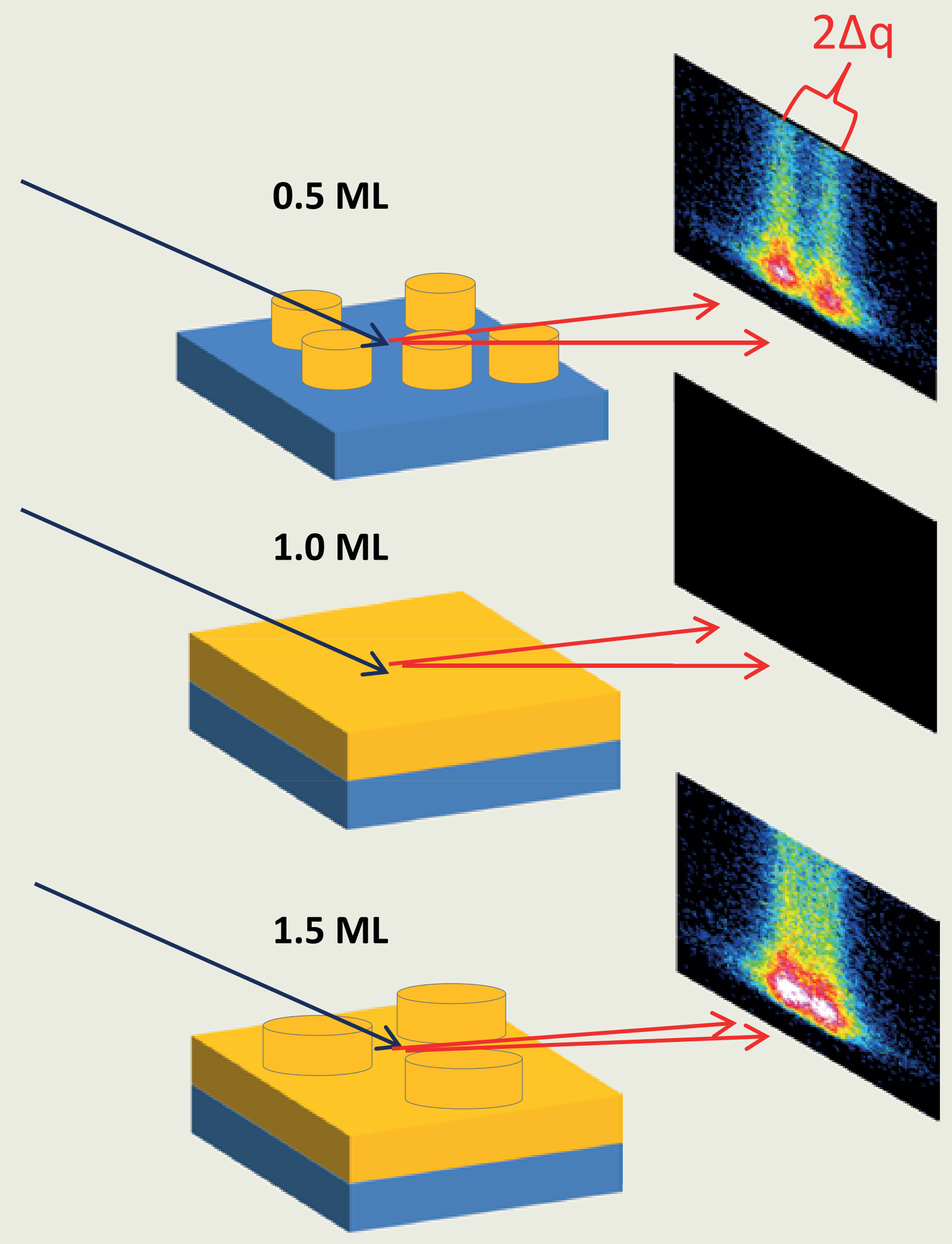
Rate equation model layer growth
(V. Trofimov et al.)

$$\frac{d\theta_n}{dt} = \begin{cases} R_1(1-\theta_1) + R(\theta_1 - \xi_n) & ,n = 1 \\ R(\xi_1 - \xi_n) & ,n > 1 \end{cases}$$

$$\xi_n = \begin{cases} 0 & ,\theta_n < \theta_{n,cr} \\ 1 - \exp\left[-\left(\sqrt{-\ln(1-\theta_n)} - \sqrt{-\ln(1-\theta_{n,cr})}\right)^2\right] & ,\theta_n \geq \theta_{n,cr} \end{cases}$$

$\theta_{n,cr}$: *Critical Coverage*

Diffuse: Island Density Evolution



- Island density

$$N = \left(\frac{\Delta q}{2\pi}\right)^2$$

(assuming square lattice)

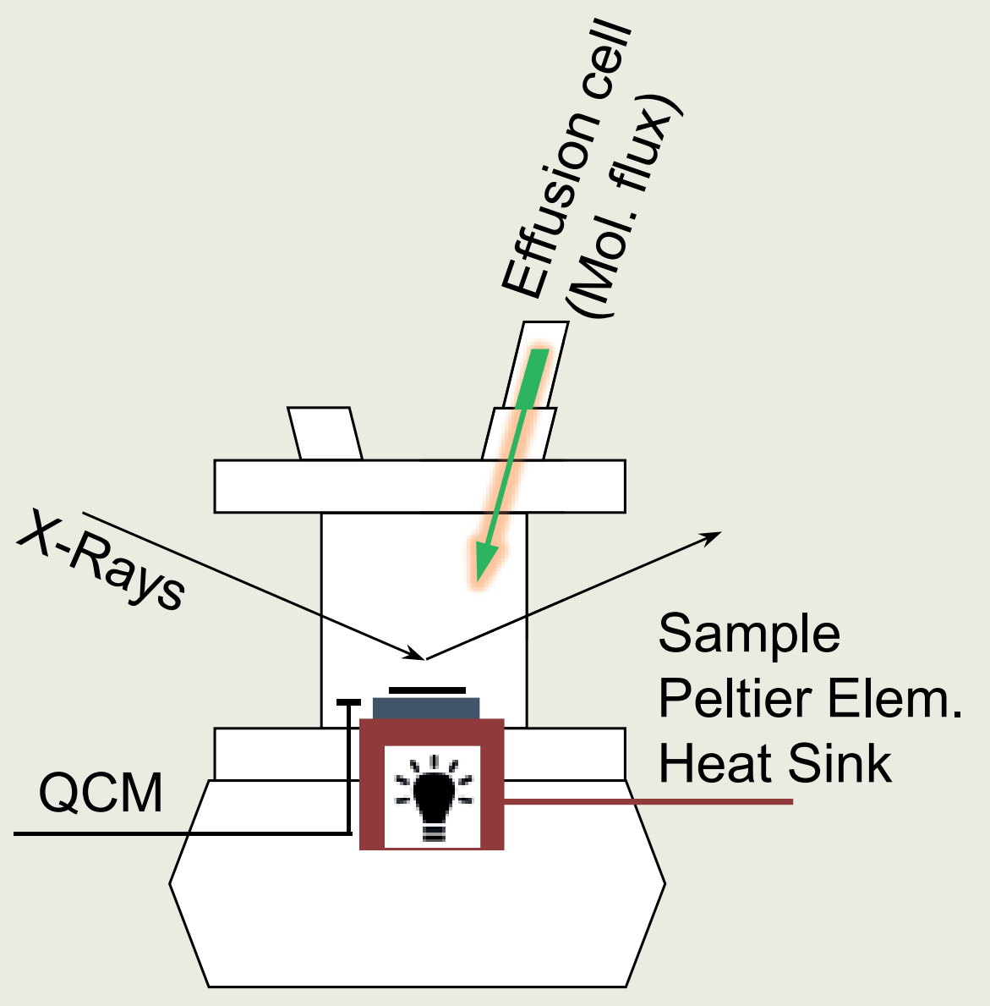
- The evolution of the island density distribution during the nucleation of molecular island can be followed *in situ* and in real-time
- For smooth, fully filled monolayers the signal intensity of the diffuse scattered signal drops completely.
- With the nucleation of islands in succeeding layers the diffuse scattered signal revives again.

Exploring new strategies to control thin film growth

In this experiment (ID10, 14keV, 0.62° incidence angle) we aimed for especially flat and homogenous organic thin films as demanded for device relevant applications.

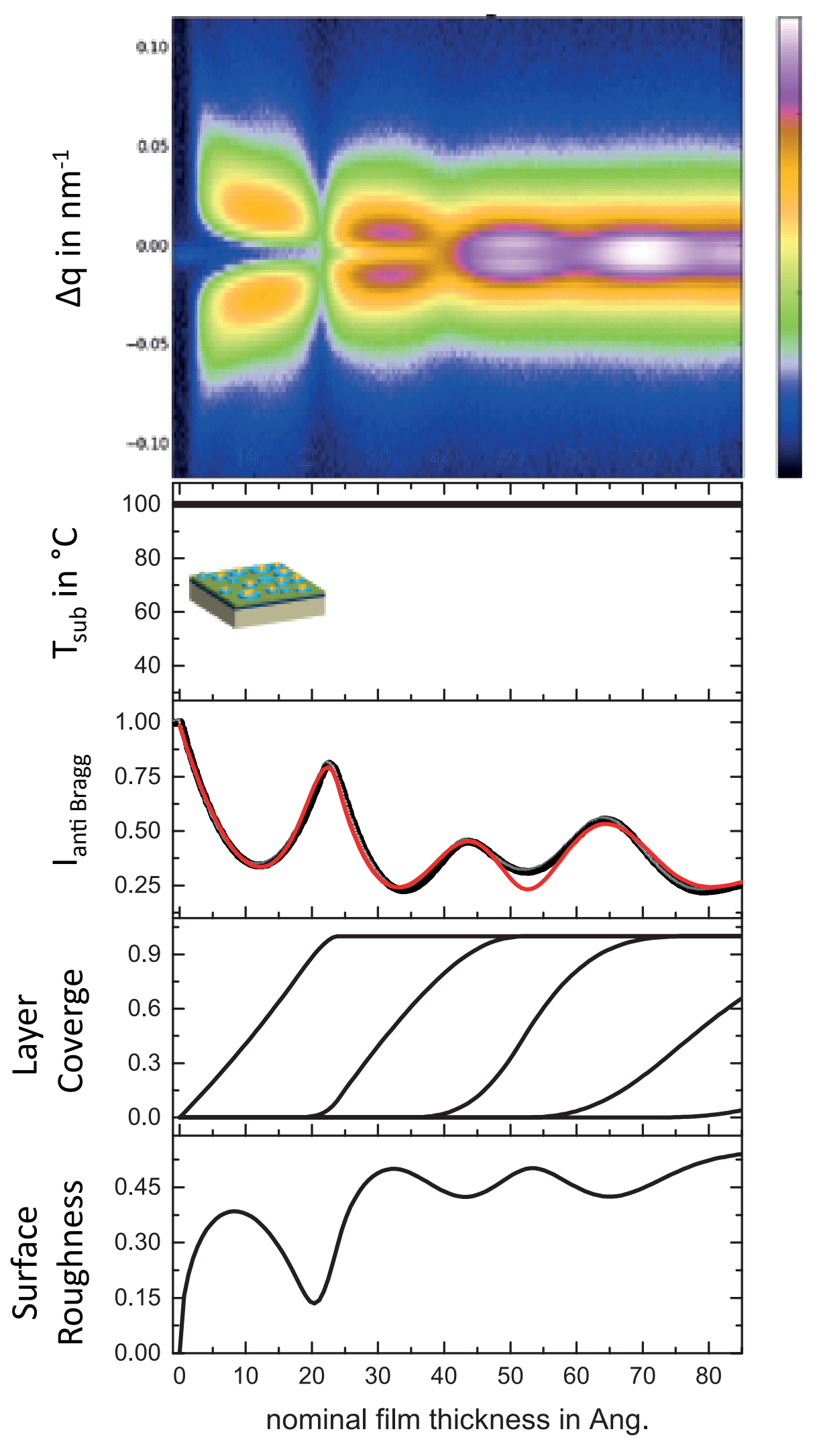
With increasing film thickness molecular films tend to roughen and to grow in a 3d fasion rather than in a layer-by-layer mode.

In order to grow high quality, smooth organic thin films we employed rapid cooling cycles during the early stages of the growth of each monolayer. This results in an increased nucleation density while preserving high molecular diffusivities at higher temperatures for the rest of the time.

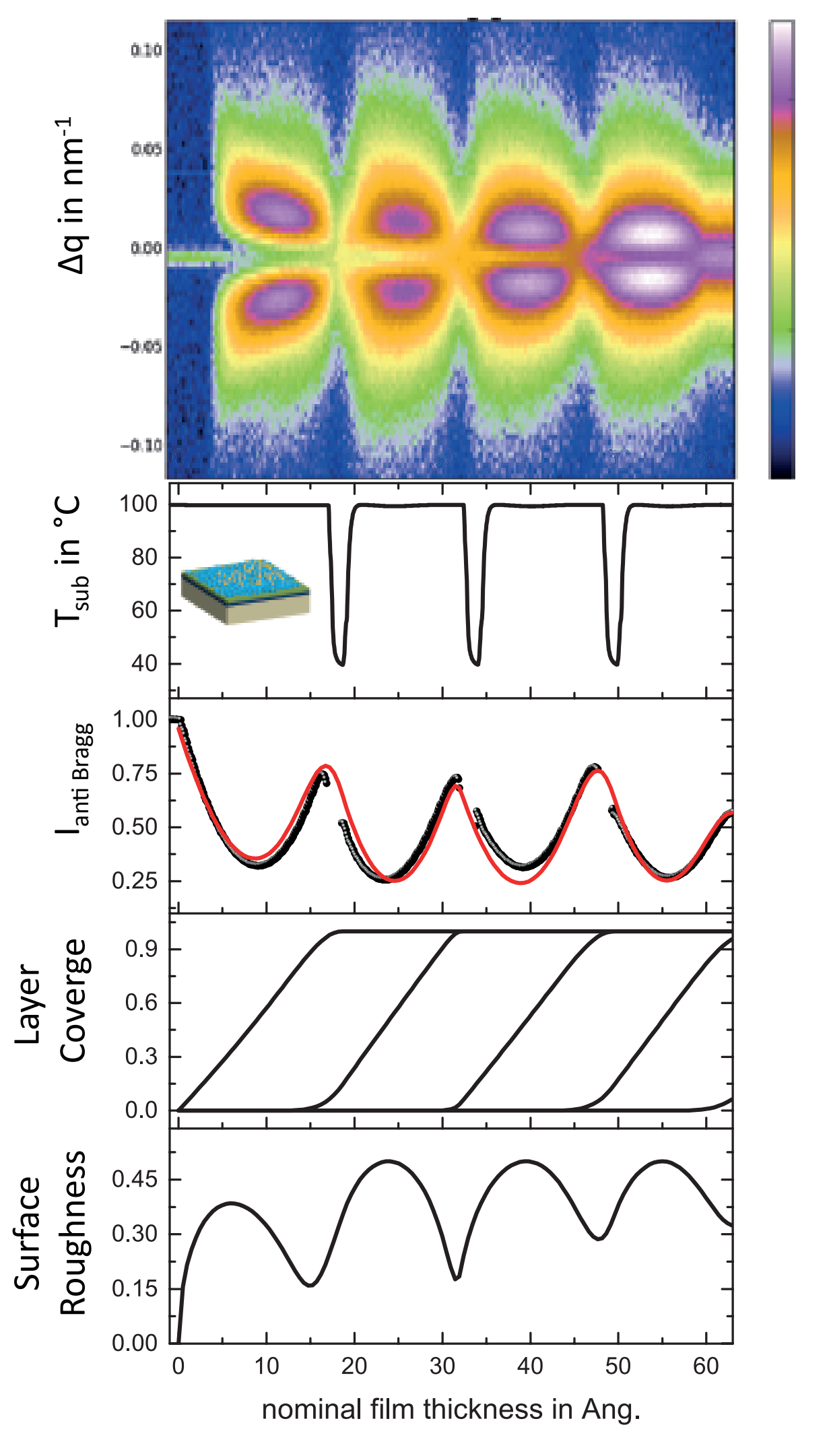


- Evaluation of the anti Bragg oscillations reveals that the growth mode is strongly shifted towards layer-by-layer growth through the cooling cycles

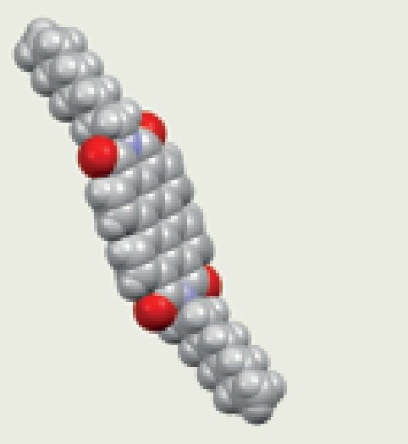
Classical thin film growth:
fixed substrate temperature



New approach:
Rapid T_{sub} cycles



- Looking at GISAXS-splitting (resolving up to 650nm large structures) over time we find that rapid cooling cycles strongly increase the nucleation density during growth in multilayered films
- This *in situ* real-time control mechanism is suitable to produce organic thin films of superior quality



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