EUROPEAN SYNCHROTRON RADIATION FACILITY

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ESRF

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office via the User Portal:

https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Reports supporting requests for additional beam time

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Crystallization phenomena in 3D DNA superstructure crystals with guest molecules	**Experiment number**: SC-4609
Beamline: ID02	**Date of experiment**: from: 30.08.2017 to: 01.09.2017	**Date of report**: 28.11.2017
Shifts: 6	**Local contact(s)**: Alessandro MARIANI	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

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Report:

In our 6 shifts of beam time at ID02 we investigated DNA origami superstructure crystals including guest gold nanoparticles with SAXS. In addition, we studied DNA origami objects freely dispersed in aqueous solution and DNA-containing lipid nanoparticles. Prior to the experiment we assembled DNA origami crystals with guest particles of different size in annealing reactions. The lattice polymerization of two precursor solutions, kept at room temperature and 47 °C, respectively, was studied in situ.

We used an X-ray energy of 12.4 keV, a beam size of 0.2x0.3 mm^2 and a sample-to-detector distance (SDD) of 3 m. For selected samples we recorded additional detector images at 1 m and 10 m SDD. The sample environment was a 1.5 mm quartz glass flow-through cell kept at 23°C by a Peltier thermostat coupled to a 40°C water bath. To study the disassembly of the DNA origami crystals and their monomeric units when heating, the samples were exposed in situ to temperature ramps up to 78°C. The in situ polymerization experiments of precursor solutions as well as the lipid nanoparticle studies were done in 2 mm quartz glass capillaries (Hilgenberg , Malsfeld, Germany). A Rayonix MX-170HS CCD detector was used with a binning of 960x960 pixels of 0.177 mm size. All samples showed isotropic scattering so the radially averaged intensity curves provided in real time were used for further analysis.

As preliminary experiments, water, TAE buffer containing $MgCl_2$, 10 nm gold nanoparticle stock solution (EM.GC10, BBI International) and rectangular DNA origami blocks were measured to choose the exposure times. Typical exposure times ranged between 10x0.05s for gold-containing samples prone to aggregation upon X-ray exposure and 10x0.5s for bare DNA constructs.

Firstly, bare DNA origami crystals and their corresponding monomers were measured and molten by heating ramps. The lattice structure determined by the molecular design was

confirmed. It was found that melting occurs just above the optimal assembly temperature. DNA origami lattices with 10 and 20 nm guest gold nanoparticles as well as gold nanorods were measured, molten and compared to SAXS intensities of the individual components.

Secondly, lipid nanoparticles containing single- and double-stranded DNA were measured. Thirdly, freely dispersed DNA origami objects with different guest particle geometries were investigated and confirmed previous particle distance measurements.

In regular time intervals throughout the whole experiment the two precursor mixtures for DNA origami crystals at 47°C and room temperature were placed in the beam and crystal formation was observed. The resulting assembly quality was better at 47°C.

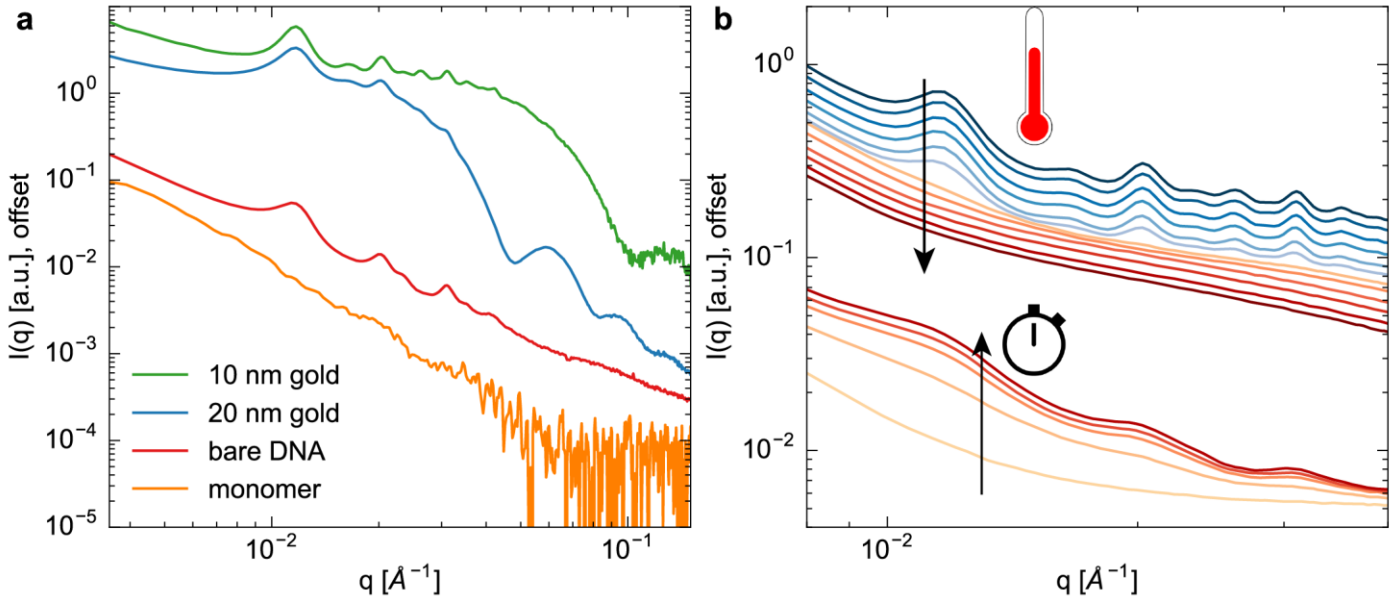

Figure 1: a) SAXS intensities of selected crystalline DNA assemblies and the monomeric unit. b) Top: Melting series of a 10 nm gold-decorated DNA origami crystal with increasing temperature, bottom: Polymerization series of a pure DNA origami crystal at 47°C over time.

A detailed analysis of the obtained SAXS intensities of the DNA origami crystals, their melts, and their components will allow a better understanding of the DNA crystal formation and disintegration. Due to the low background of the flow-through cell and the high q resolution crystallite sizes and lattice parameters can be derived without additional assumptions about the shape of the components. The extracted characteristic temperatures of melting and growth serve to optimize future design protocols of DNA origami crystal monomers.