LARMOR Soft Matter

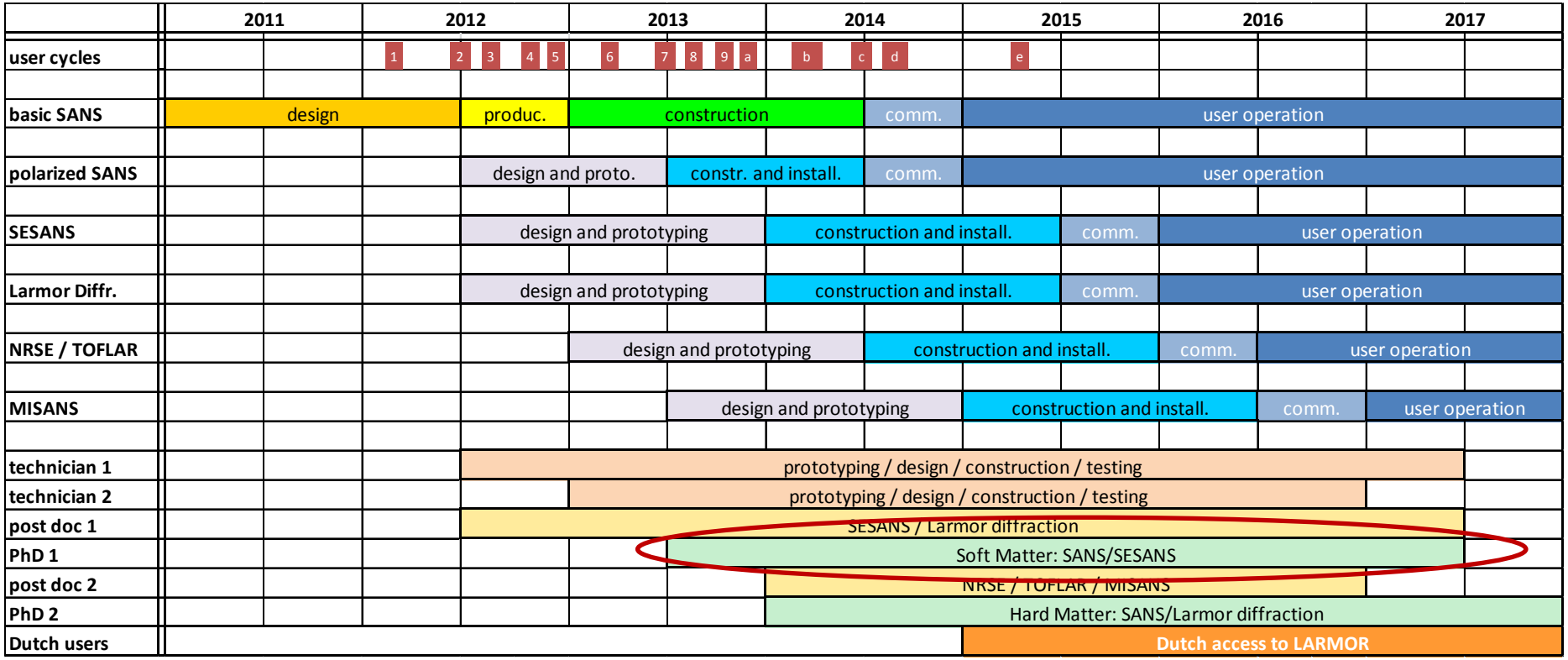
Measuring Structure and Dynamics of Self-assembling Soft Condensed Matter

Jurrian H. Bakker

Outline

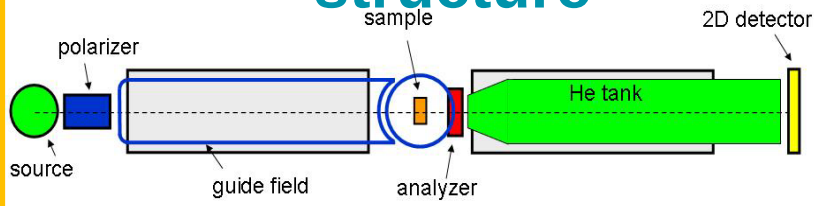
- LARMOR project
  - Soft Matter PhD
- First experiments with SESANS
  - Casein aggregation
- Modelling
  - MD simulations
- Eindhoven collaboration
  - Complex Coacervate Core Micelles (**C3M**s)
  - Benzene-1,3,5-tricarboxamides (**BTA**s)

LARMOR timeline

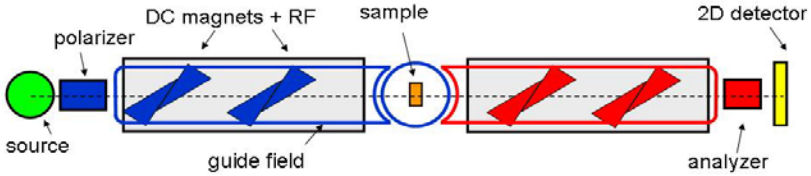


LARMOR options

structure

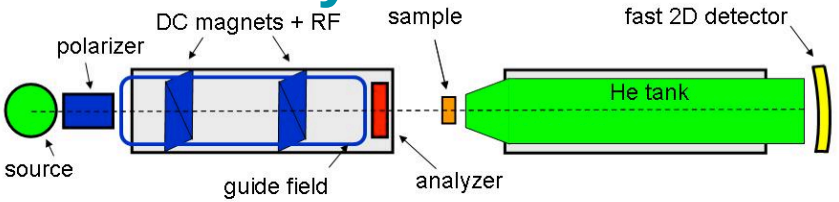


Small Angle Neutron Scattering (SANS)

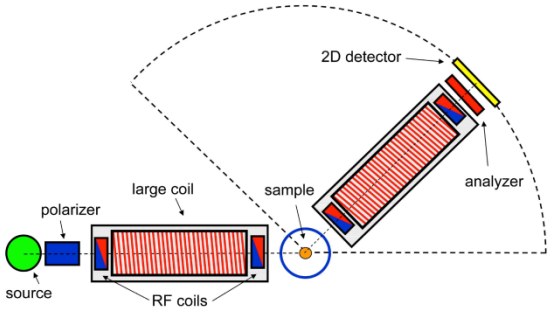


Spin Echo Small Angle Neutron Scattering (SESANS)

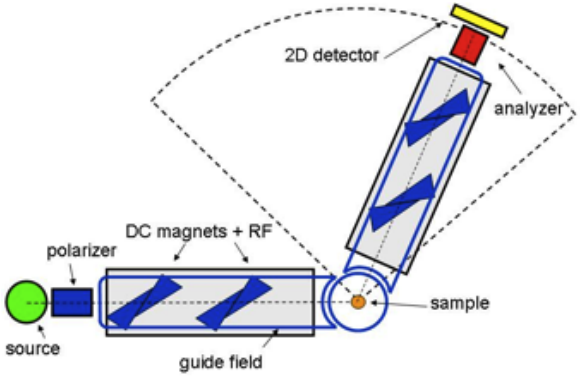
dynamics



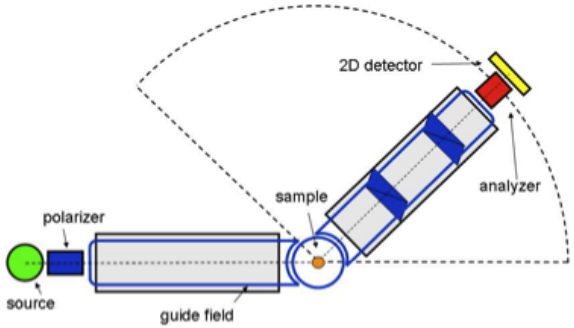
Modulated Intensity Small Angle Neutron Scattering (MISANS)



Neutron Resonance Spin Echo (NRSE)



Larmor diffraction



Time-of-Flight Larmor labeling (TOFLAR)

PhD description

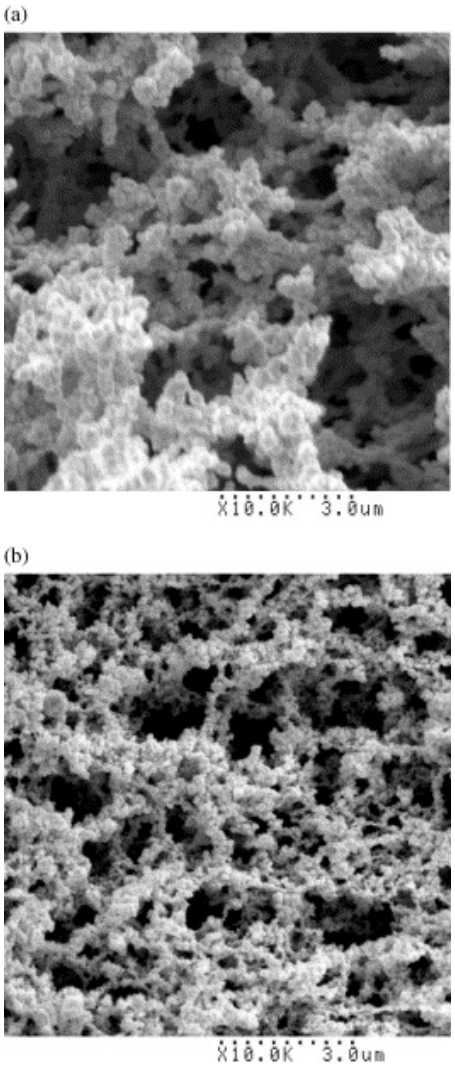
- To develop hardware and software tools for LARMOR in the field of Soft Matter
  - Hardware: sample environment, instrument geometry
  - Software: data reduction and analysis, accessible to non-expert users (biologists, chemists, material scientists)

- To use LARMOR to generate new scientific knowledge in the area of Soft (Condensed) Matter
  - Self-assembling molecular/colloidal structures

First SESANS experiments

Casein aggregation

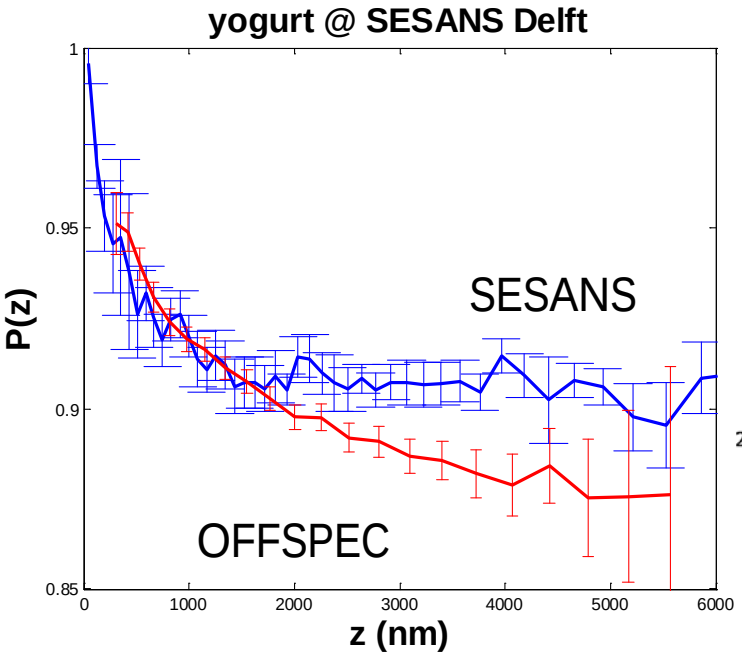
- Under acidification (pH~4.5-5), milk becomes yogurt (gelation)
- Analytical models of gelation do not fit the data
- Neither do Lattice-based Monte Carlo simulations with adhesive spheres
- So, we will use Off-Lattice Molecular Dynamics with adhesive-repulsive spheres to better approximate and characterize these structures

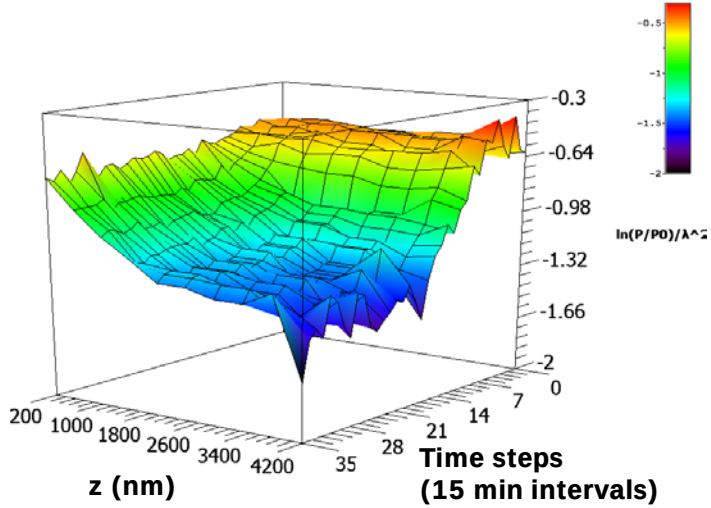


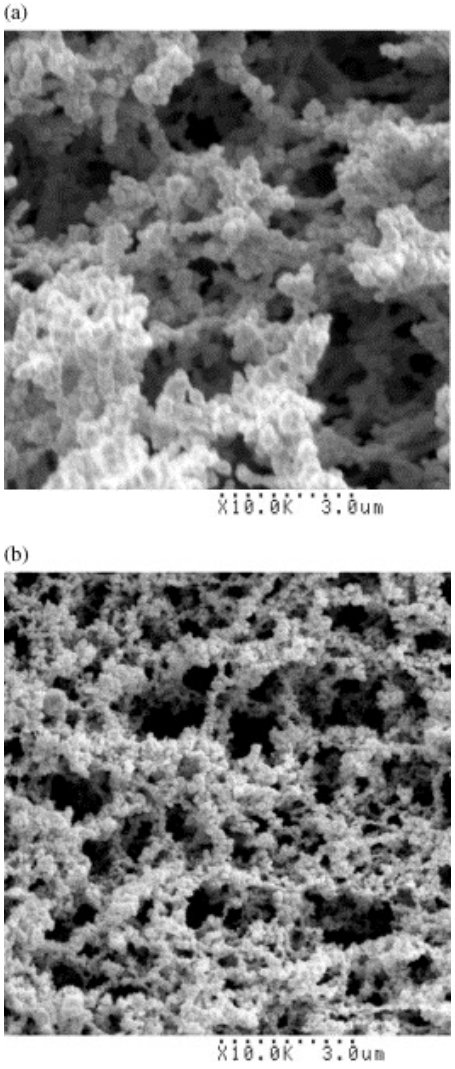
First SESANS experiments

Casein aggregation

- SESANS (Delft & OFFSPEC)
- OFFSPEC Aggregation kinetics

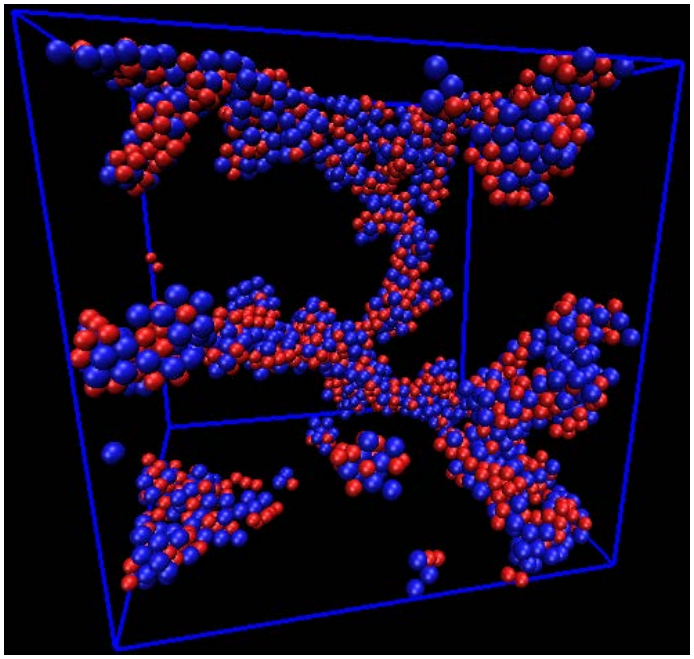


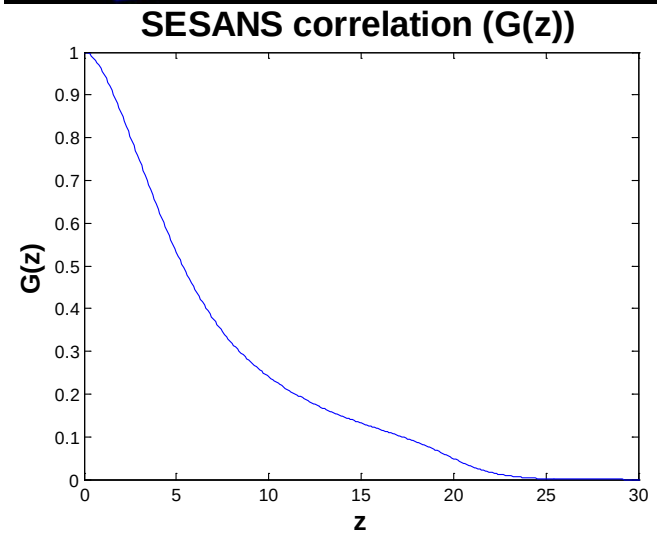




Modelling collaboration

- Collaboration with Prof. G. Foffi (Laboratoire de Physique des Solides, Université Paris-Sud)
- This will allow description of casein aggregate structures
- The collaboration will be on-going to model other experimental data





New algorithm to calculate SESANS from local colloidal structures

- Can we describe local structure of colloids with SESANS?
- Using the **Debye sum** for spherically averaged scattering

$$\mathrm{I}(Q) = \sum_{j=1}^{n}\sum_{k=1}^{n}\left(f_j(Q)\cdot f_k(Q)\cdot\frac{\sin(Q\cdot|r_j-r_k|)}{Q\cdot|r_j-r_k|}\right)$$

- If we assume all scatterers to be identical spheres of radius R we can write

$$S(Q) = \sum_{j=1}^{n}\sum_{k=1}^{n}\left(\frac{\sin(Q\cdot|r_j-r_k|)}{Q\cdot|r_j-r_k|}\right)$$

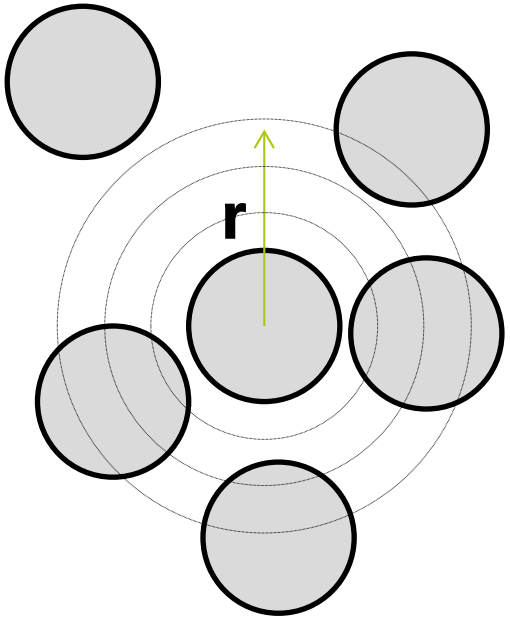
$$P(Q) = \left(\frac{3\cdot[\sin QR - QR\cdot\cos QR]}{(QR)^3}\right)^2$$

- (inverse) spherical Fourier transform of I(Q) gives the **auto-correlation** function γ(r)

$$\gamma(r) = \frac{1}{2\,\pi^2}\int_0^\infty I(Q)\cdot\frac{\sin(Q\cdot\Delta r)}{Q\cdot\Delta r}\cdot Q^2\,dQ$$

- Hankel transform of I(Q) gives the **projected correlation function** G(z) (SESANS)

$$\mathrm{G}(z) = \frac{\int_0^\infty J_0(Q\cdot z)\cdot\ I(Q)\cdot Q\ \ dQ}{\int_0^\infty I(Q)\cdot Q\ \ dQ}$$



Future work

Scientific research and LARMOR development

- Scientific work
  - With ICMS group @ TU/e and prof. Foffi @ U-PSud
- SESANS data analysis
  - Development of Sasview software
- Larmor sample environment
  - Control of pressure, temperature, humidity,...
  - Critical for soft matter experiments

Soft matter collaboration @ LARMOR

- NPM2 (Neutron and Positron Methods in Materials) provides SANS and SESANS options (and expertise)

- MSc J.H. (Jurrian) Bakker
- Dr. W.G. (Wim) Bouwman
- Dr. A.A. (Ad) v. Well
- Prof. dr. C. (Catherine) Pappas

- ICMS (Institute for Complex Molecular Systems) provides chemical synthesis and samples (and expertise)

- Dr. I.K. (Ilja) Voets
- Dr. N. (Neus) Vilanova Garcia
- (MSc I. (Isja) de Feijter)

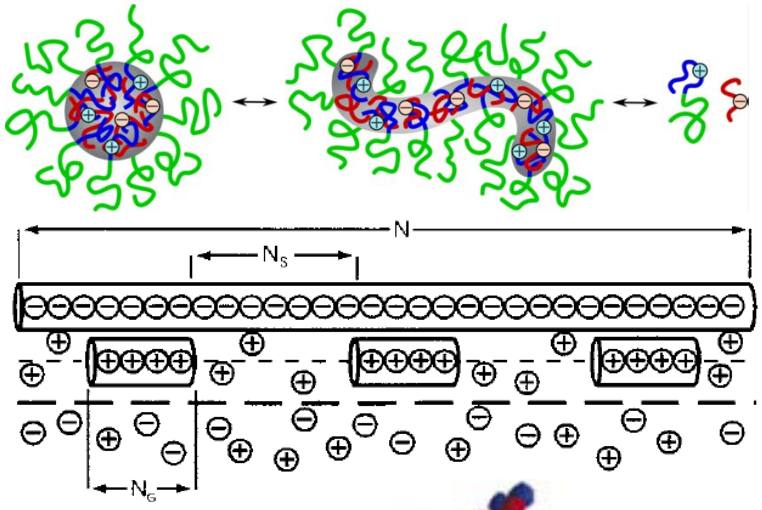
Gene therapy delivery system

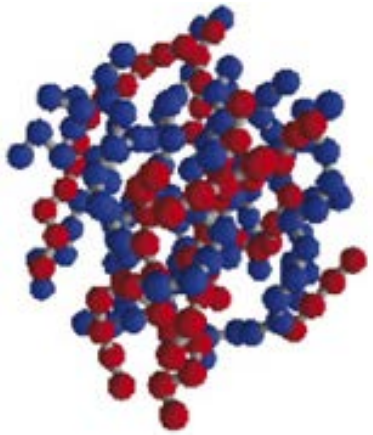
Complex Coacervate Core Micelles (C3Ms)

- **Self-assemble** readily from components at room temperature
  - Mix of poly-anions and poly-cations

    → **Phase separation**
  - If one poly-ion is co-polymerized

    → **Micro-phase formation**





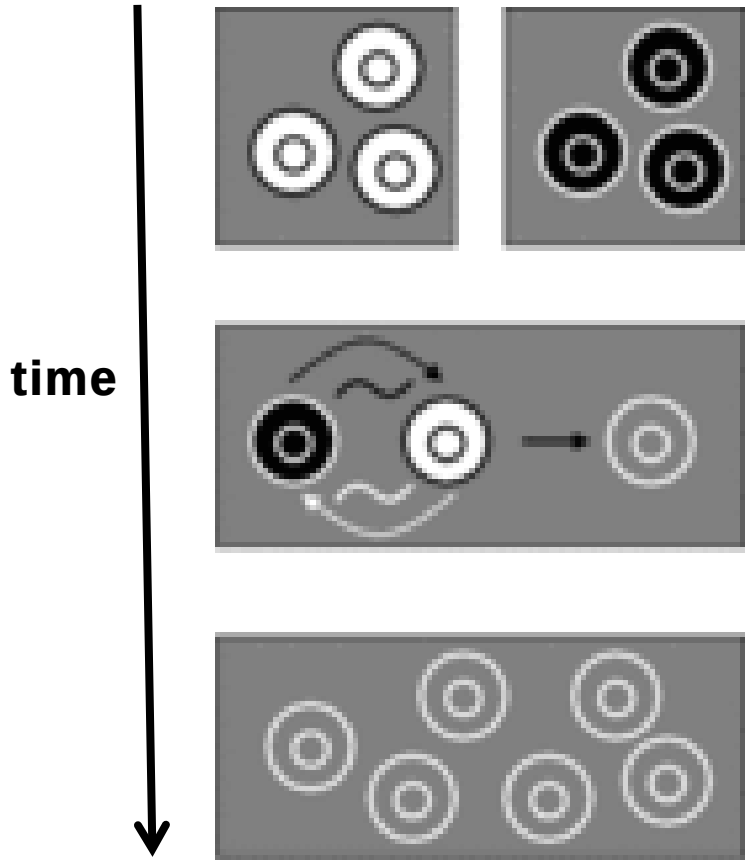
- Applications:
  - Wastewater purification, Nano-reactors
  - Drug delivery, Chelation, Controlled release
  - **Non-viral Gene therapy**
- Remaining questions
  - Equilibrium chain exchange kinetics are hardly known

    → **Important for gene therapy**

C3Ms – Proposals submitted

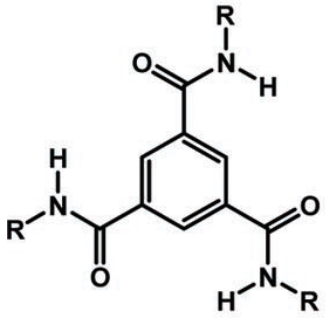
Time Resolved SANS Contrast decay experiments

- ISIS (UK) & LLB (France)
  - Spallation and reactor experiments to compare results from different and complementary instruments
  - Mixing H and D micelles and observe loss of contrast in time, as well as where that loss occurs in the micelles

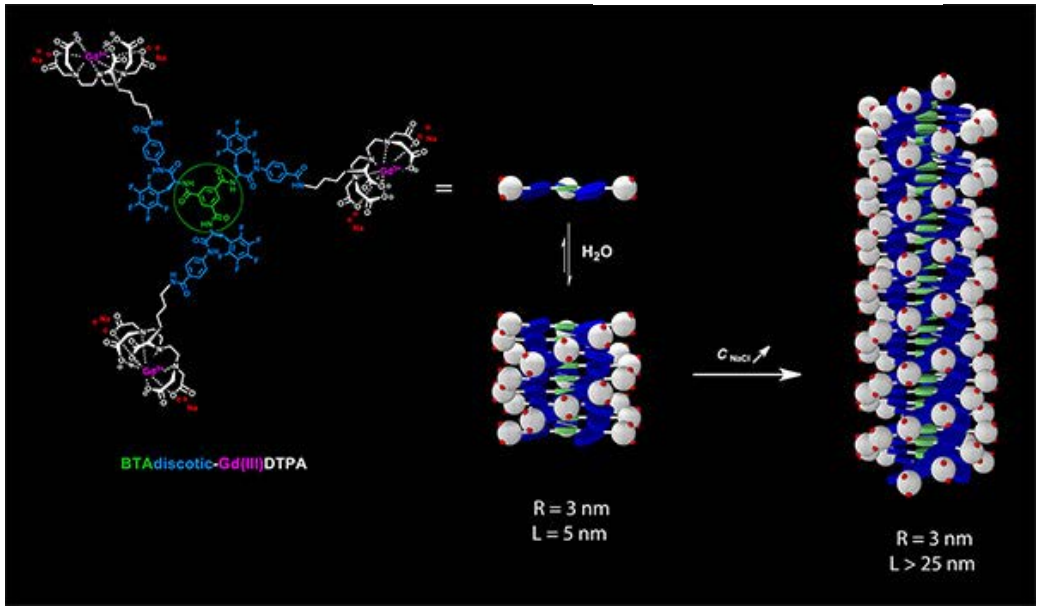


BTA – Tuneable connectors

SANS experiment for exchange rate



- Benzene-1,3,5-tricarboxamide
- Self-assembling helical supra-molecules with variable pitch and chemistry
  - Chemistry will control self-assembly w.r.t. light, temperature (pH, ions)
  - Applications for electronics, biomedical engineering, sensors,...



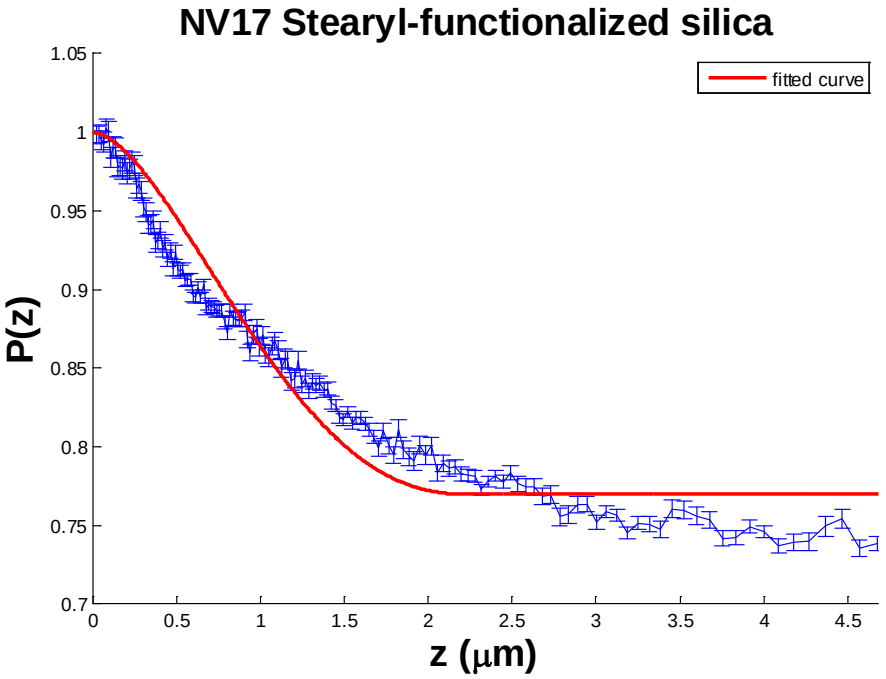
Exchange of monomers between helical assemblies (stability) is unknown
→ contrast decay experiment

This will be a later project

BTA-Silica colloids

SESANS experiment for interaction potential

- Functionalized silica
  - Attach BTA to silica colloids and study interaction potential under light, temperature
  - Interaction potential determines stability of chiral assemblies
  - Can be done with SESANS



Some preliminary measurements done. Will be a later project.