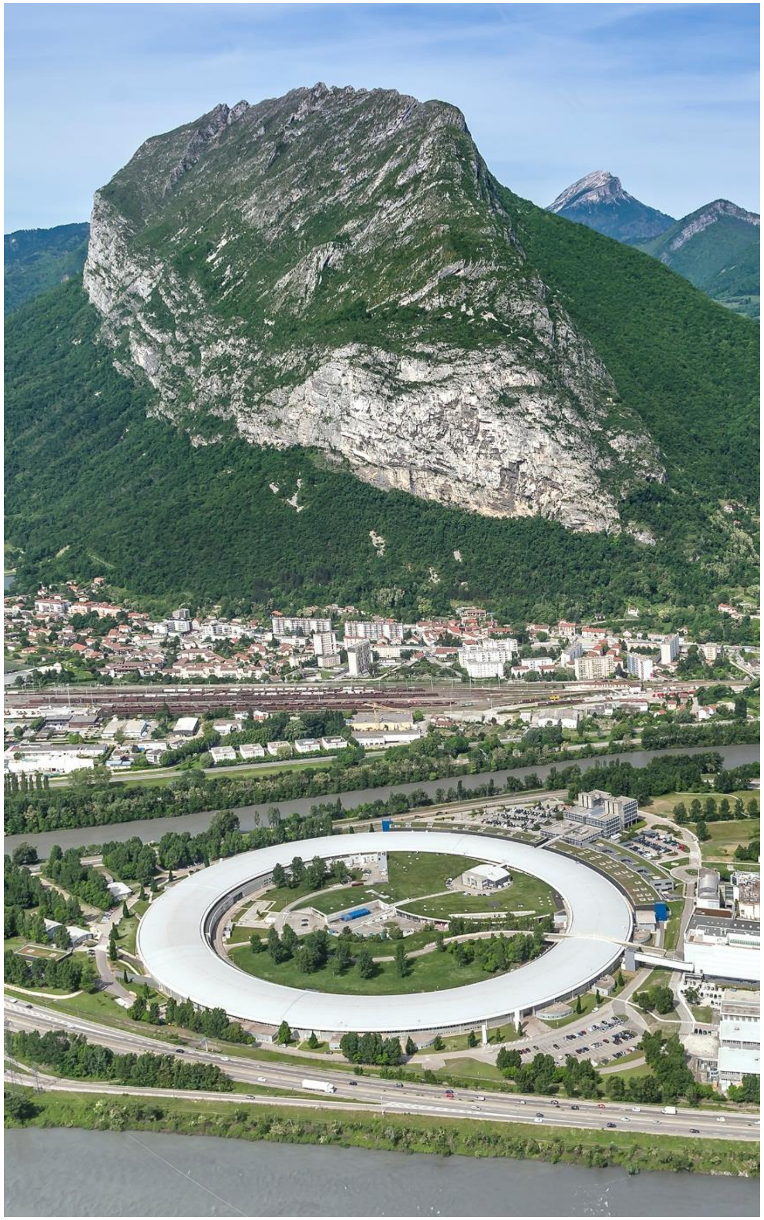
ESRF | The European Synchrotron



Perspectives for 3D Coherent X-ray Diffraction Imaging at the ESRF EBS

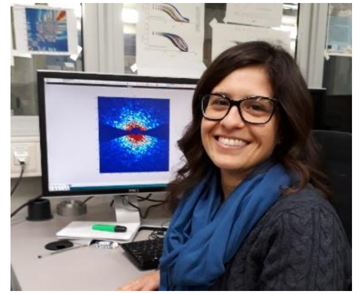
Federico Zontone

ID10 - Complex Systems and Biomedical Science group

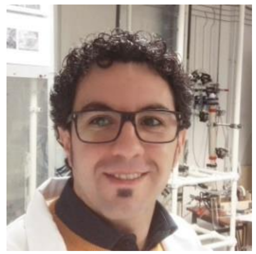
ESRF – The European Synchrotron

… the fellows…

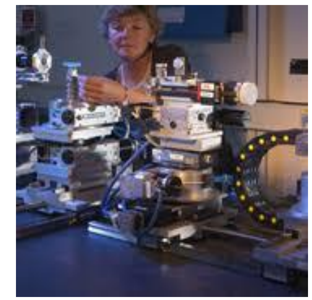
Yuriy Chushkin
ID10-CBS, ESRF



Carla Polo
Visitor from LNSL,
Campinas, Brazil



Salvatore Sanzaro



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Giovanni Mannino

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Thomas Beuvier

Prof. Alain Gibaud

Outline

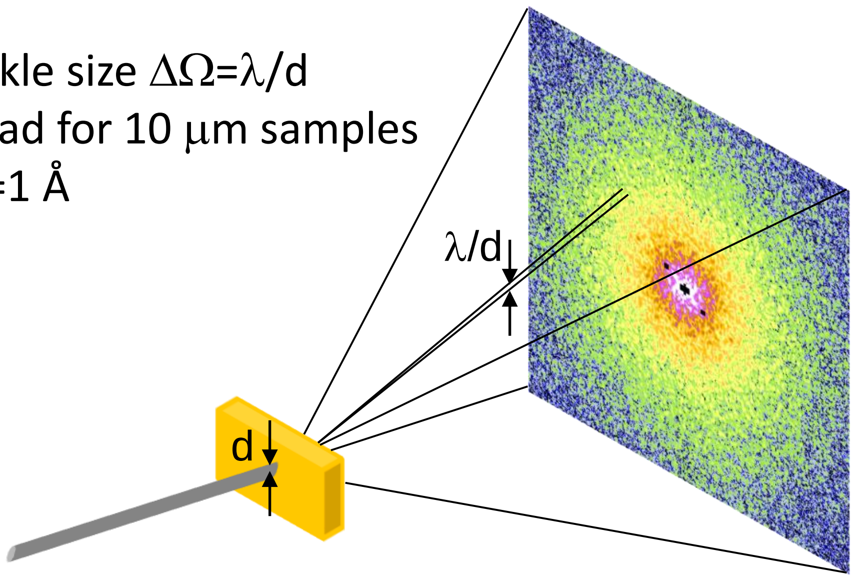
- CXDI: why and how
- 3D CXDI at ID10CS
- Examples of applications at ID10CS
- 3D CXDI at ID10CS: status and outlook
- 3D CXDI and EBS L1

CXDI: why and how

An object can be imaged by looking at **SPECKLES**

Speckle pattern: **interference pattern in the far-field** when $V_{scatt} \leq V_{coh} = \xi^2_T * \xi_L$

Speckle size $\Delta\Omega = \lambda/d$
10^{-5}rad for 10 μm samples
at λ=1 Å



$$I(q,t) \propto \left| \sum_n f_n(q) \cdot e^{iq \cdot r_n(t)} \right|^2$$

Speckles encode the **exact spatial frequencies** of the (disordered) system

High-resolution (q_{max}) 3D imaging ($\rho_e(\mathbf{r})$) can be obtained by simple inverse Fourier transform

CXDI: why and how

Lost phases can be (numerically) retrieved if speckles are **oversampled** (sampled better than Nyquist frequency, **$\sigma > 2^{1/N}$**)

Iterative phase retrieval algorithm

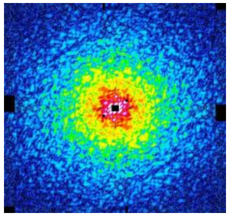
Initial estimate

$\rho(r)$

FFT[$\rho(\mathbf{r})$]

$$F(q) = |F(q)|\, e^{-i\Phi(q)}$$

Measured |F(q)|



Support, positivity

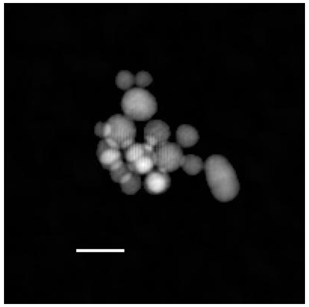
$$F'(q) = \sqrt{|I(q)|}\, e^{-i\Phi(q)}$$

Add random Φ

FFT^{-1}[F($\mathbf{q}$)]

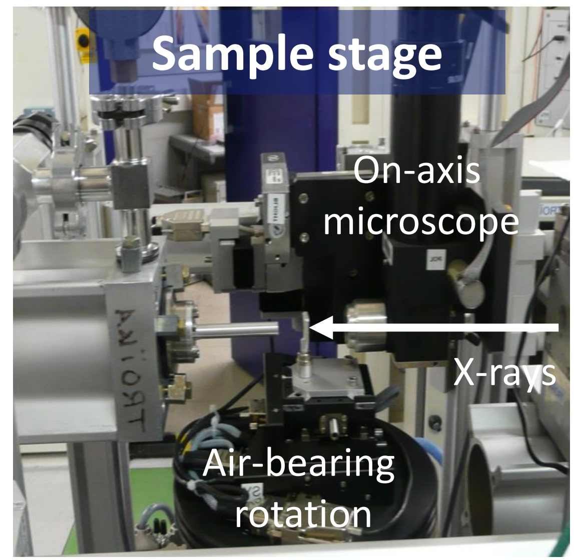
$\rho'(\mathbf{r})$

Final output



3D CXDI at ID10

Tomography setup for samples in air and SAXS geometry

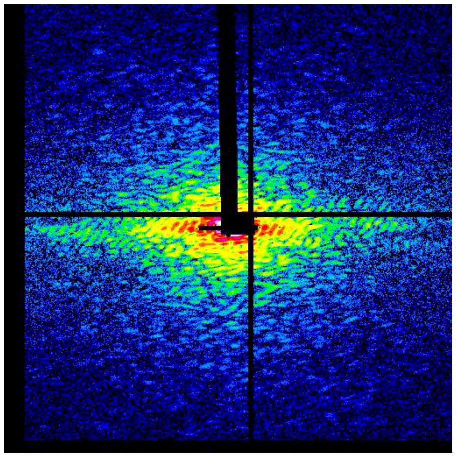


10 µm beam
$7x10^{10}$ ph/s at 8.1 keV
Samples on Si_3N_4 membranes
Maxipix2x2 (516x516, 55 µm pixel size)
Detectors at max 7m from sample

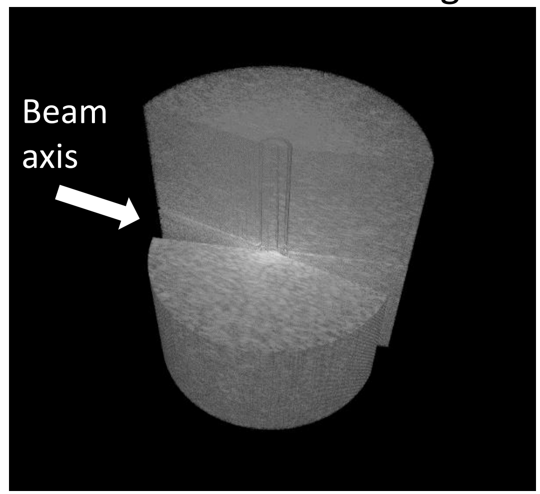
3D CXDI at ID10

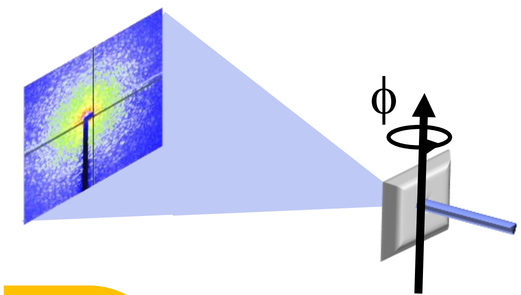
From data collection…

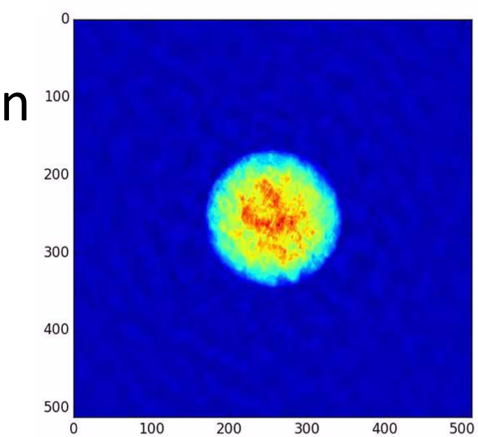
ϕ scan: -84°,+78°, 0.25° step



512x512x512 Fourier grid





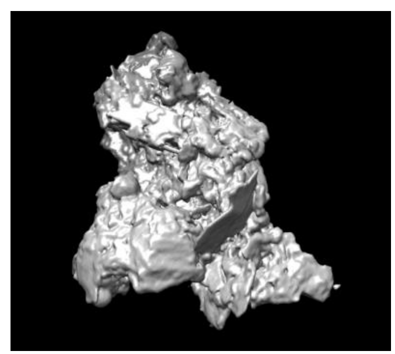


… to sample reconstruction

Phase retrieval algorithm applied directly to 3D Fourier space (3D FFT):

- HIO and ER
- shrink-wrap for support

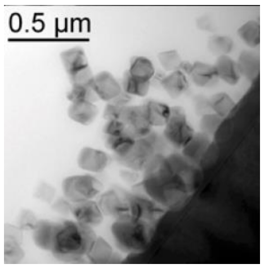
Yuriy’s in-house software



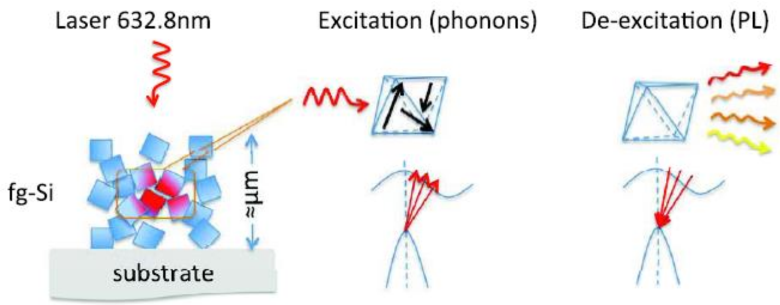
Chushkin, Zontone et al., J. Synchrotron Rad. 21(2014)594

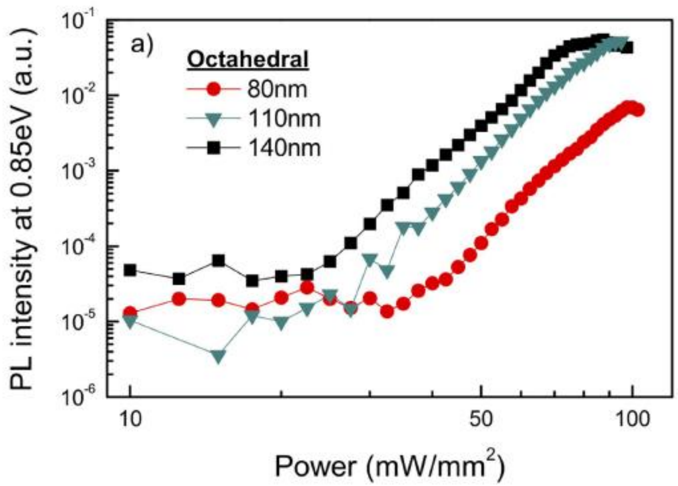
Meso-structured semiconductors

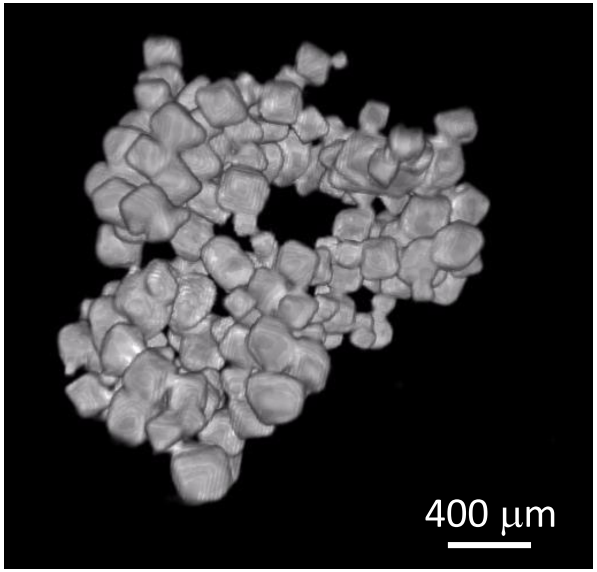
Stimulated giant photoluminescence by phonon confinement in faceted 100 nm Si crystals



TEM of Si grains, obtained by inductively coupled plasma CVD





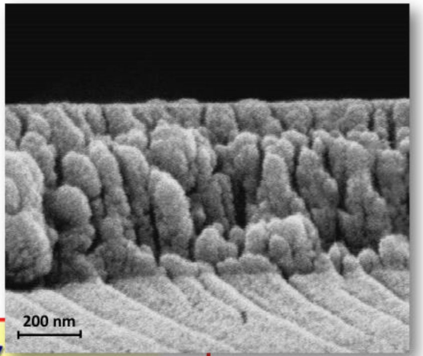


3D CXDI shows that assemblies of large octahedral grains favor the largest PL yield (highest porosity, lowest heat dissipation)

de Jong et al., Sci. Rep. 6, 25664 (2016)

Nano-engineered TiO_2 materials

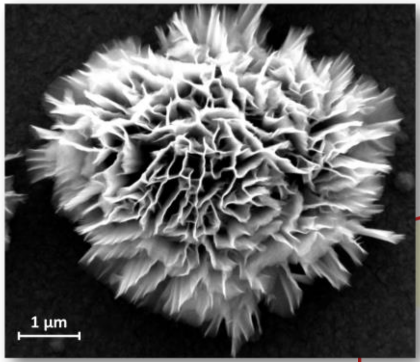
TiO_2 nano-rods deposited by reactive sputtering in grazing incidence geometry assisted by local oxidation (GIG-LOX)

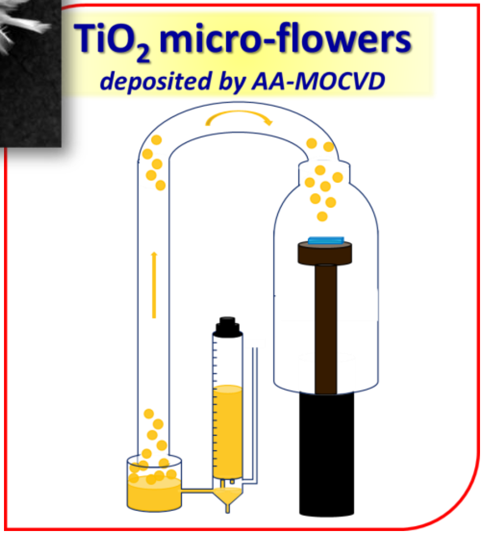


Sanzaro et al., Sci. Rep. 6, 39509

vs.

TiO_2 micro-flowers deposited by Aerosol Assisted Metal-Organic Chemical Vapor Deposition (AA-MOCVD)



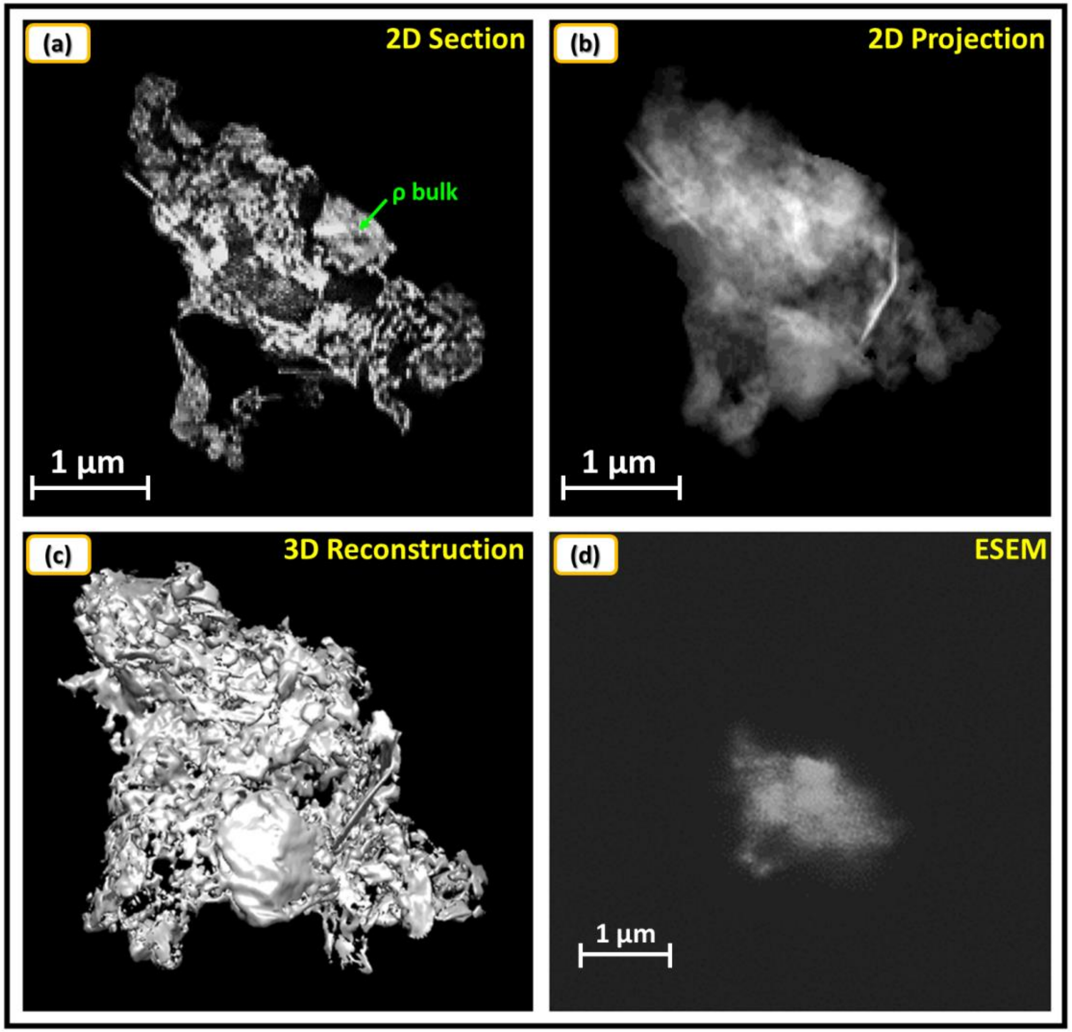


Salvatore Sanzaro
PhD thesis
Messina University (Italy)

<u>Supervisor:</u> *Dr. Alessandra Alberti, IMM - Catania (Italy)*

<u>Supervisor:</u> *Dr. David Muñoz-Rojas LMPG - Grenoble MINATEC (France)*

Nano-engineered TiO_2 materials

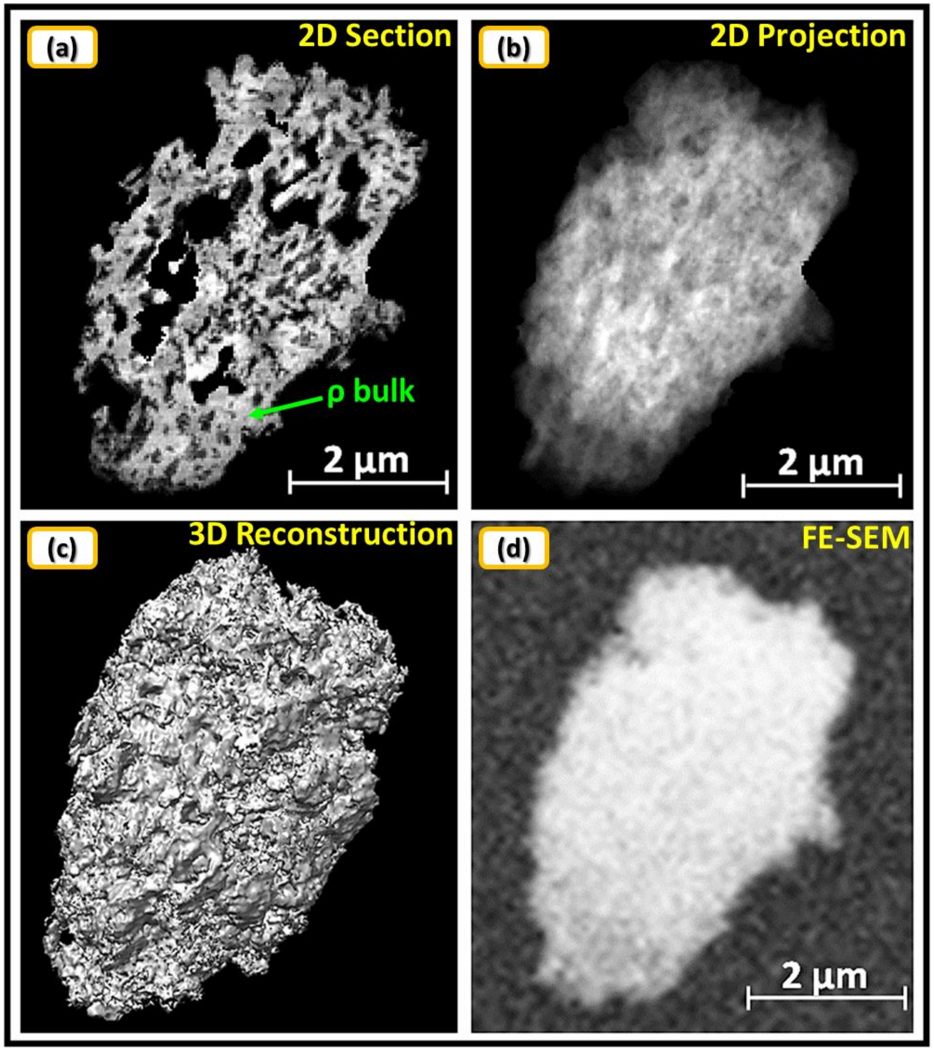


TiO_2 gig-lox layer

(a) Central slice
(b) Volume representation
(c) 3D reconstruction by CXDI
(d) ESEM image

Surface-to-Volume ratio = **0.06 nm^{-1}** (by CHIMERA software)

Nano-engineered TiO_2 materials



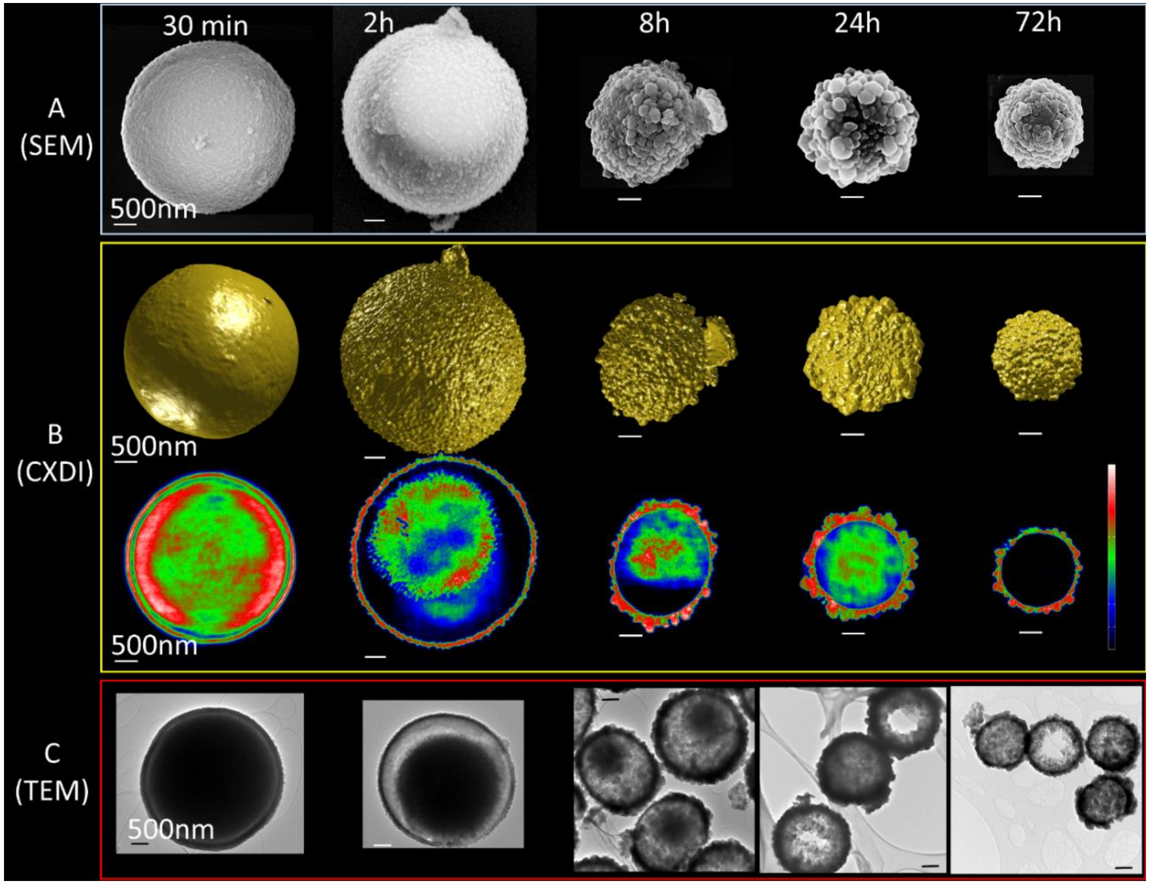
TiO_2 micro-flower

(a) Central slice
(b) Volume representation
(c) 3D reconstruction by CXDI
(d) FE-SEM image

Surface-to-Volume ratio = **0.04 nm^{-1}** (by CHIMERA software)

Bio-mimetic materials and crystallization processes

Following the formation of hollow $CaCO_3$ microsphere from (Ca^{2+}), (CO_3^{2-}) solutions with a polystyrene sulfonate molecule



- Amorphous CC nucleates and forms porous vaterite that rapidly transforms into non-porous calcite

- With PSS vaterite crystallization is inhibited: a thin shell forms, more stable than the core

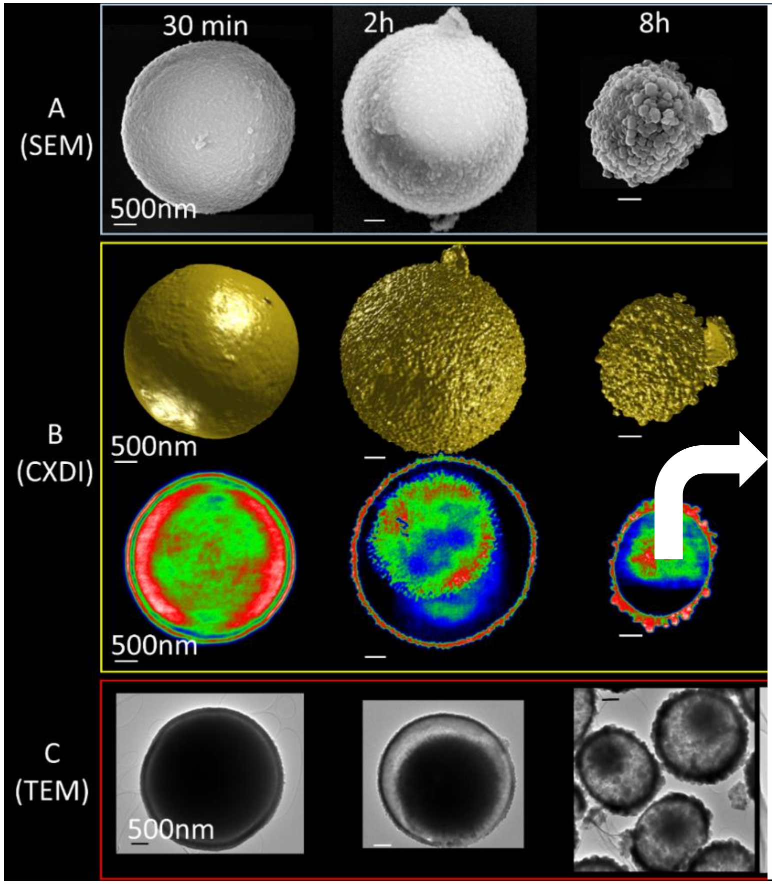
- With time, small vaterite crystals form outward the shell, while particles shrink and inner cores dissolve

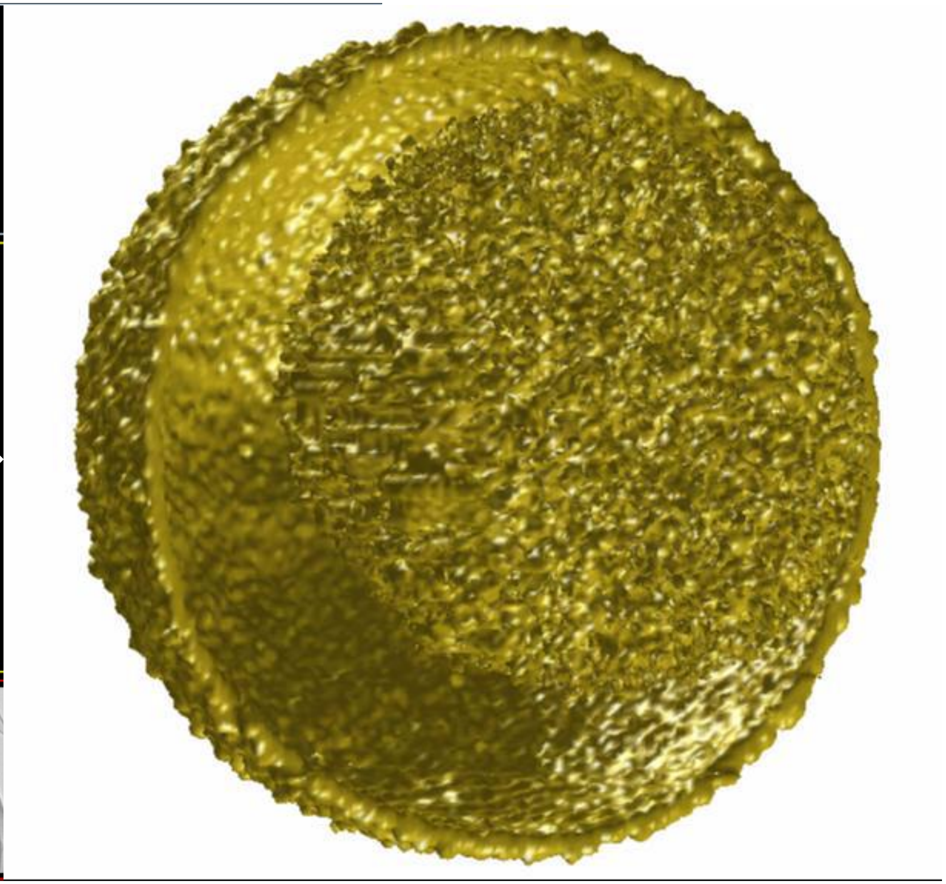
SEM/TEM perfectly match CXDI reconstructions but can not probe the inside (particles are too big)

Thomas Beuvier at ID10

Bio-mimetic materials and crystallization processes

Following the formation of hollow $CaCO_3$ microsphere from (Ca^{2+}), (CO_3^{2-}) solutions with a polystyrene sulfonate molecule



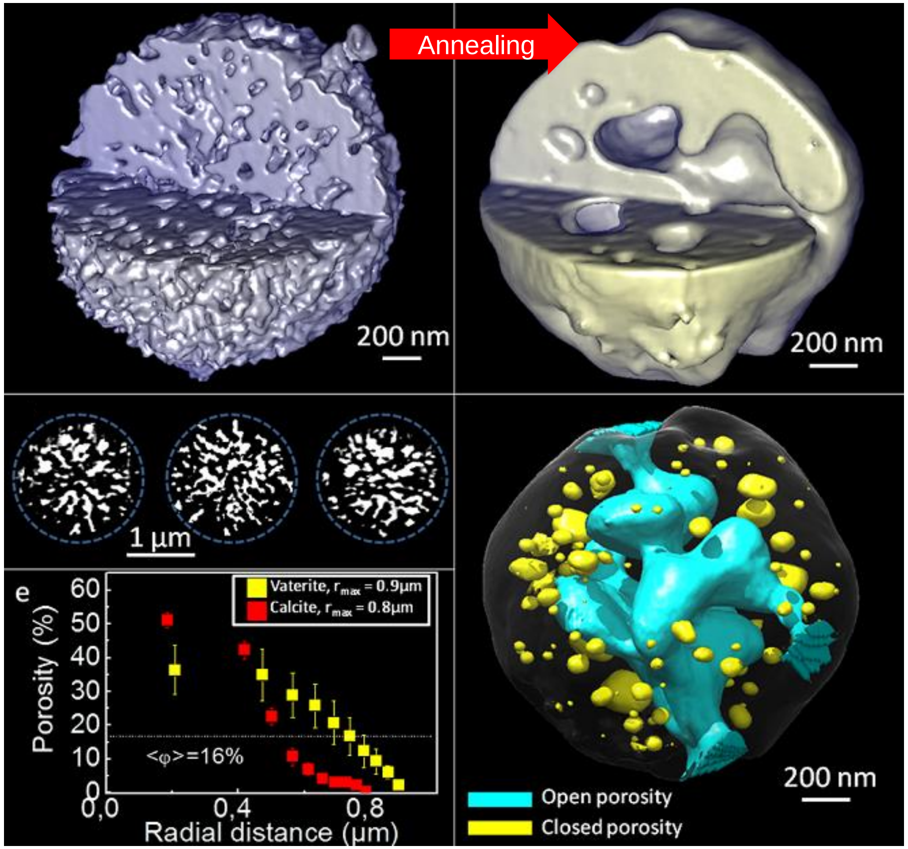


SEM/TEM perfectly match CXDI reconstructions but can not probe the inside (particles are too big)

Thomas Beuvier at ID10

Bio-mimetic materials and crystallization processes

Porosity evolution in microspheres during vaterite-to-calcite solid-state transformation by annealing at 420 °C



- Pore geometry totally changed

- Mainly open and spherical pores in Calcite microspheres

- Constant total pore volume (~16%)

- Specific surface is substantially reduced in Calcite (from 7.3(8) to 2.9(2) m^2/g)

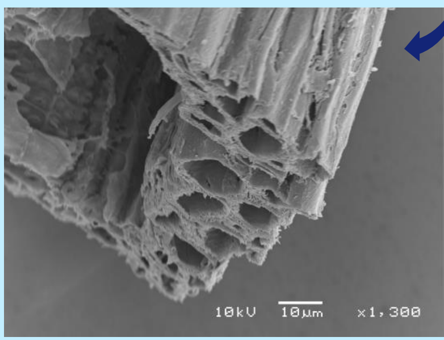
- The change in the pore morphology is governed by surface free energy minimization

By 3D CXDI is possible to control the internal morphology of $CaCO_3$ microparticles during a phase transformation

Cherkas et al., Cryst. Growth Des. 2017, 17, 4183

Environment: Assessment of sugarcane bagasse nano-structure

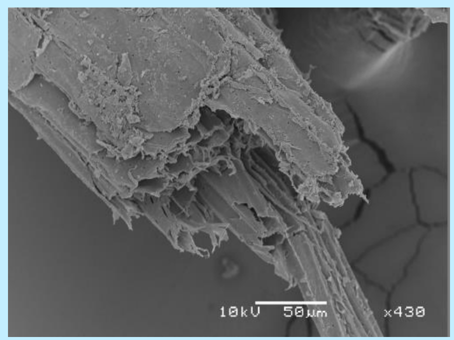
Sugarcane bagasse
from 1st generation ethanol



Lignocellulosic matrix

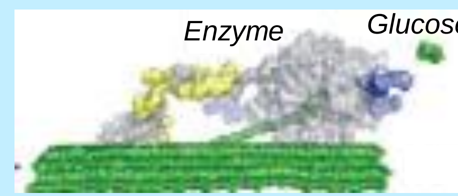
Pretreatments
Physical-chemical process

PRETREATMENT METHOD
Uncatalyzed steam explosion
Liquid hot water
pH controlled hot water
Flow-through liquid hot water
Dilute acid
Flow-through acid
Ammonia fiber expansion (AFEX)
Ammonia recycle percolation (ARP)



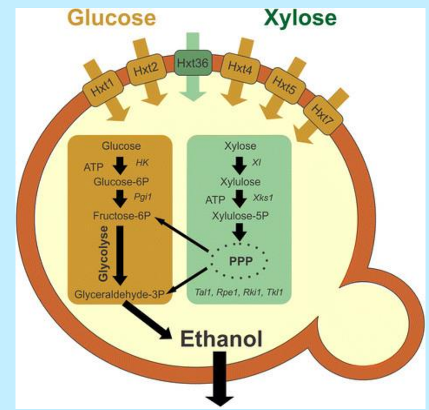
Cellulose release

Enzymatic hydrolysis
Glucose release



Cellulose microfibrils

Fermentation
Yeast



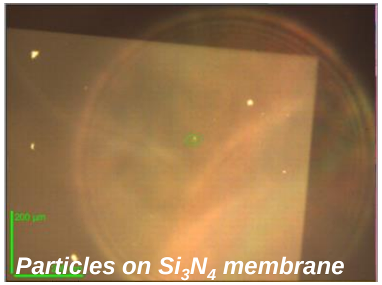
2nd generation ethanol

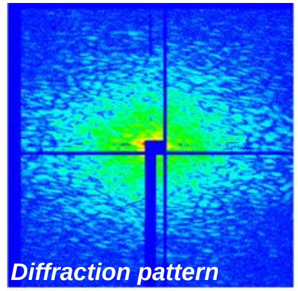
Rose et al., *Ind. Biotechnol.*, 2013/ Nijland et al., *Biotechol. Biofuels*, 2014

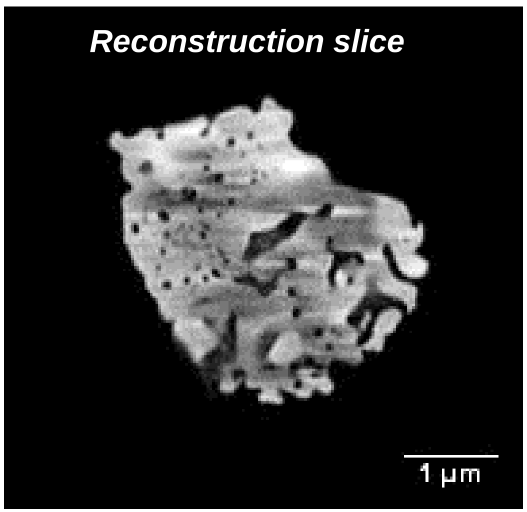
Carla Polo at ID10

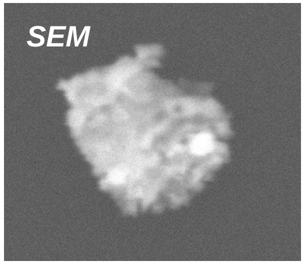
Environment: Assessment of sugarcane bagasse nano-structure

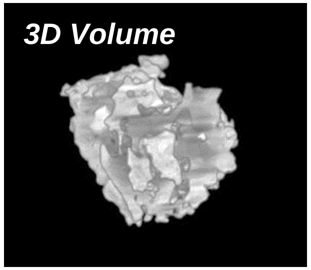
In natura Brazilian sugarcane bagasse





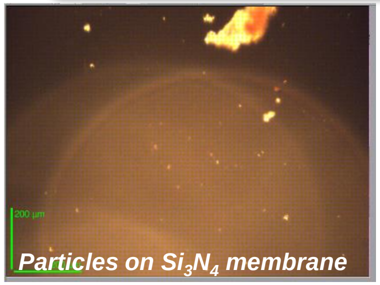


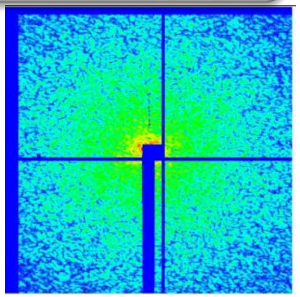


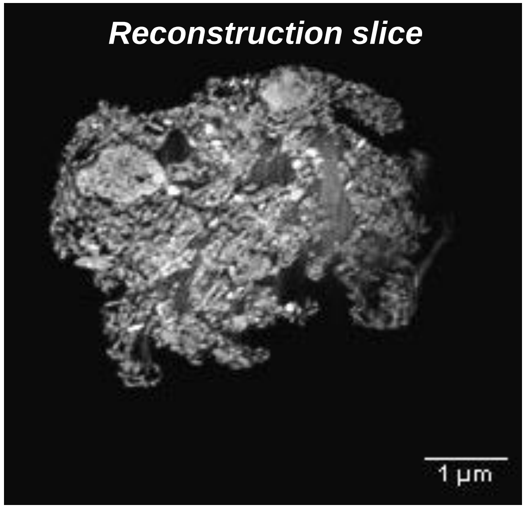


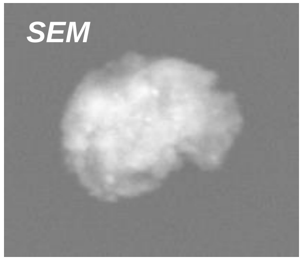
Hydrothermal pre-treatment

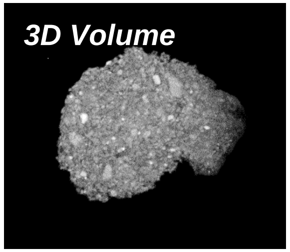
In natura Brazilian sugarcane bagasse submitted to pretreatment











3D CXDI at ID10CS: status and perspectives

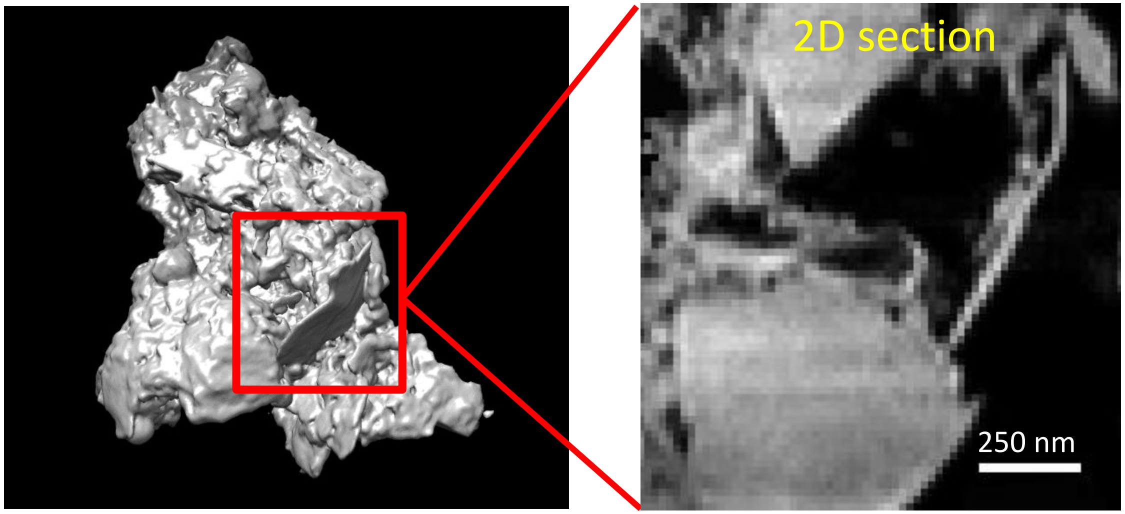
PROs

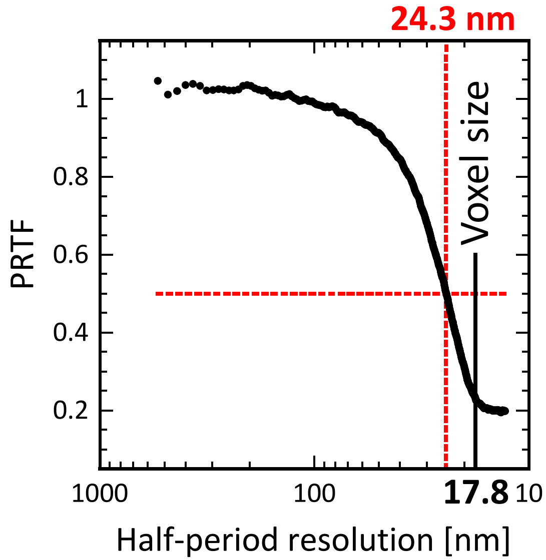
- Lens-less, full-field imaging technique
- q_{max} resolution
- Tolerant to translational instabilities
- **Well suited for tomography at high resolution**

CONs

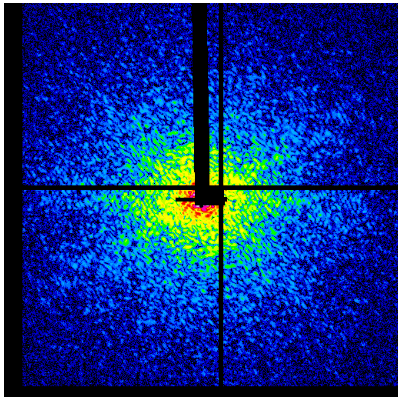
- Limited FoV (few μm)
- Needs **isolated objects**
- Slow technique (few hours)
- Reconstructed ρ(r) not (yet) in absolute units

3D CXDI@ID10CS is hitting the setup limitations





Pixellated details: **Half-period resolution close to voxel size**



Further improvement limited by attainable q_{max}

- Limited flux
- Limited focusing capabilities (10µm beam only)
- Limited detector distance (max 7m)
- Limited Maxipix detector (0.25M only)

3D CXDI at EBS L1

The EBS machine will lead to ~**100X increase in brilliance**

100X increase in coherence flux ($F_c=B\lambda^2$)

- Increase spatial resolution below 10 nm
- Acquisition times from hours to minutes

Larger transverse coherence length

- Increase FOV up to 25 μm

EBS L1 is the project **fully dedicated** to coherence in the far-field for **XPCS**, **XCCA** and **CXDI** (today, ID10CS is operating in 50% time sharing mode)

Goals

10nm resolution with >10^3 resolution elements (>10μm samples) in 3D

Go to higher energies (E > 10 keV)

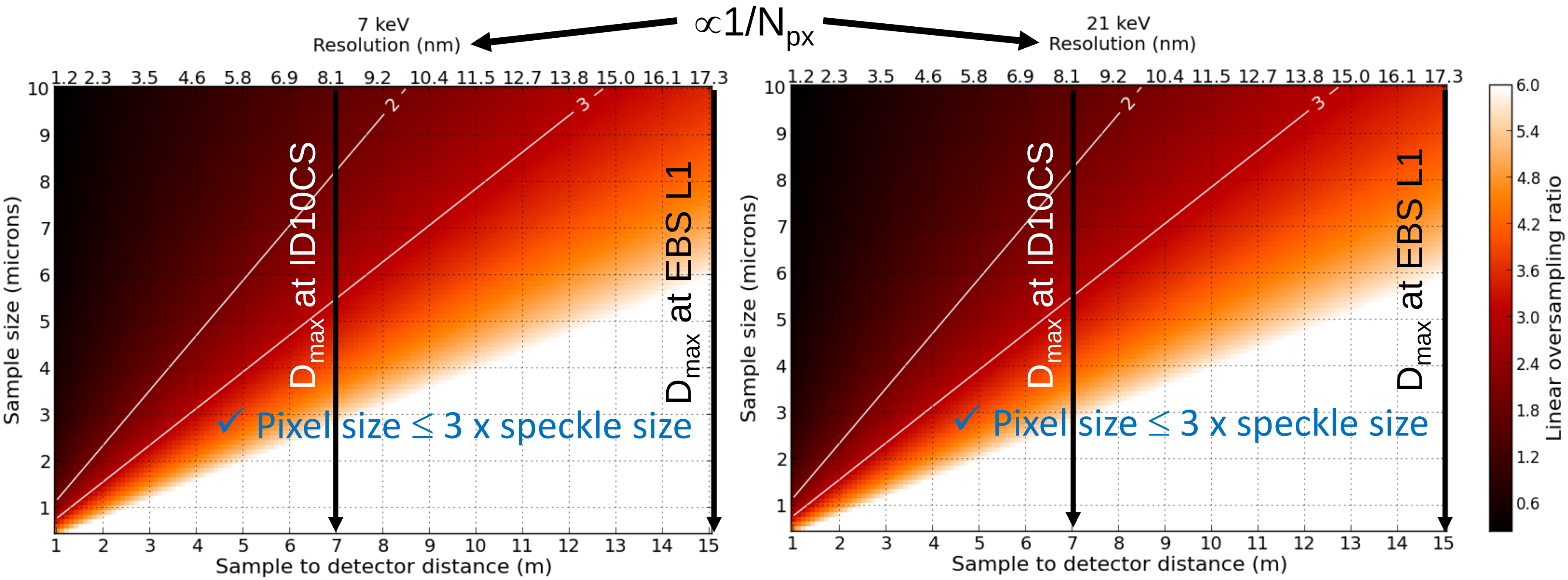
Time-resolved 3D-CXDI (*in situ, in operando* environments)

3D CXDI at EBS L1

EBS L1 promises to remove the present limitations

- 100X higher flux
- Increased focusing capabilities (down to 3µm beams)
- Increased detector distance to max 15m

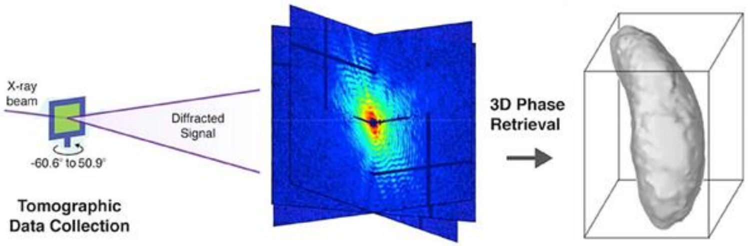
- Detectors like Eiger 4M or MÖNCH 16M (in future) will achieve the goals



Eiger, 75 µm pixels size

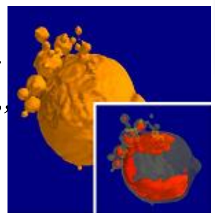
MÖNCH, 25 µm pixels size

EBS L1: Many new scientific challenges



Rodriguez et al., IUCrJ, 2015
Frozen hydrated Neospora Caninum

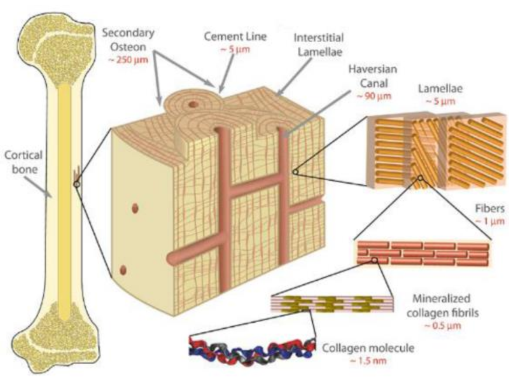
Frank et al., Sci. Rep 7, 14081,2017
Cryo-fixed malaria-infected "ghost" human RBC

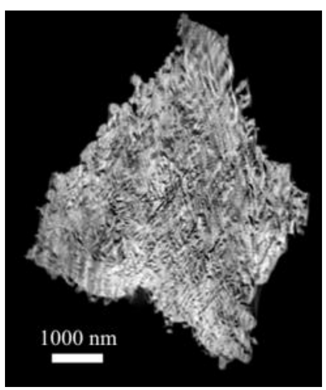


3D imaging at 5-10 nm resolution with CXDI (10 µm samples, 100x coherent flux)

- *Internal structure in cells and tissues*
- *Nano-engineered soft and hard materials*
- *Biomimetic materials and crystallization processes*

Nano-scale bone structure:
ordered arrangement of collagen molecules and mineral platelets





Zimmermann et al., in preparation
Verezhak et al., in preparation

10-100 cells per mL

Cell Type	CTC	Erythrocyte	Leukocyte
Size (µm)	12-25	5-7	7-15

Cell Type	Neutrophil	Lymphocyte	Monocyte	Eosinophil	Basophil
Size (µm)	12-15	7-10	15-25	12-15	12-15
% of Pop	60-70	20-45	2-10	1-3	0.5-1

1,000,000 cells per mL

Courtesy of C. Giannini, CNR, Bari, Italy

Thank you