

This talk is on small bubbles...



Tim Salditt

on behalf of the small-bubble collaboration

XFEL UM , HH29.1.2020

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DESY Photon Science

Johannes Möller, Jörg Hallmann, Ulrike Boesenberg, Chan Kim, Markus Scholz, Alexey Zozulya, Wei Lu, Roman Shayduk, Robert Schaffer,
Anders Madsen
MID / XFEL

- [UAC MID 06/2019 @14keV](#) and [Beamtime 10/2019 @17.5keV](#) (p2207 & p2545 Hagemann/ Salditt)

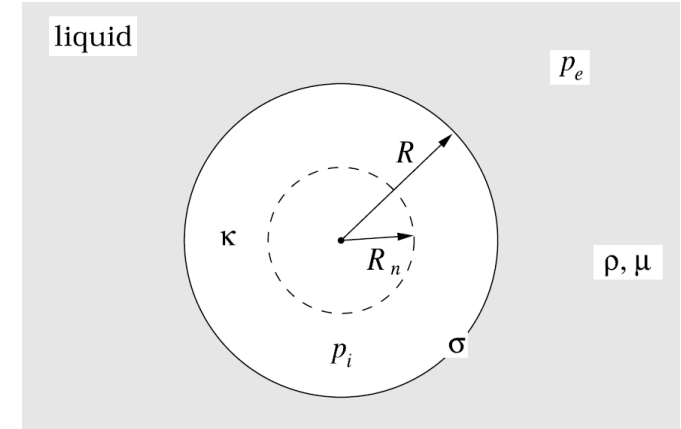
Draw me a bubble...

- (quasi-) spherical gas phase in liquid
- Cavitation bubbles
induced by hydrodynamic flow, optical breakdown, acoustic pressure
when hydrostatic pressure falls below vapor pressure
- far from equilibrium, e.g. formed by
ultrasound or optical breakdown (laser)
- **Bubbles @ ocean – atmosphere interface**

Evidence for the importance of bubbles in increasing air–sea gas flux
DM Farmer, CL McNeil, BD Johnson - Nature, 1993

Scale dependence of bubble creation mechanisms in breaking waves
GB Deane, MD Stokes - Nature, 2002

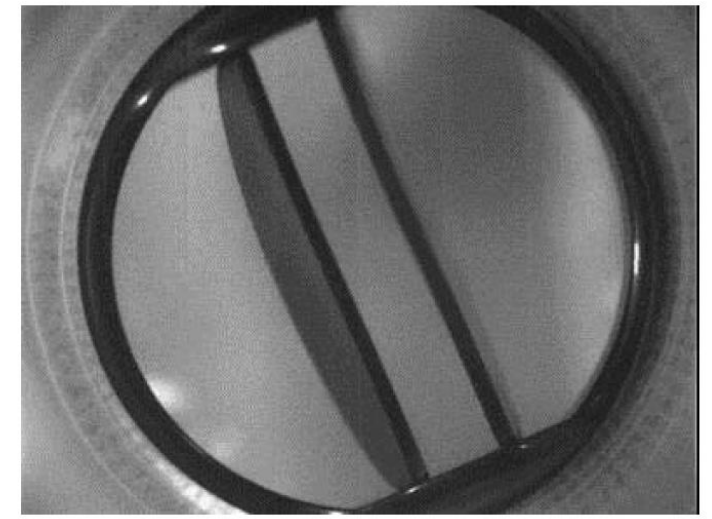
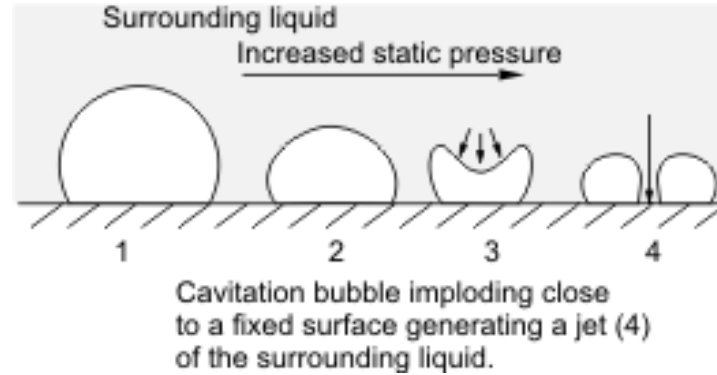
Gas transfer by breaking waves
L Deike, WK Melville - Geophysical Research Letters, 2018



Katasushika Hokusai (1760-1849)

Important in climate models !

Cavitation in engineering



CE Brennen, Interface Focus Roc. Soc. 2015

artificial heart valves

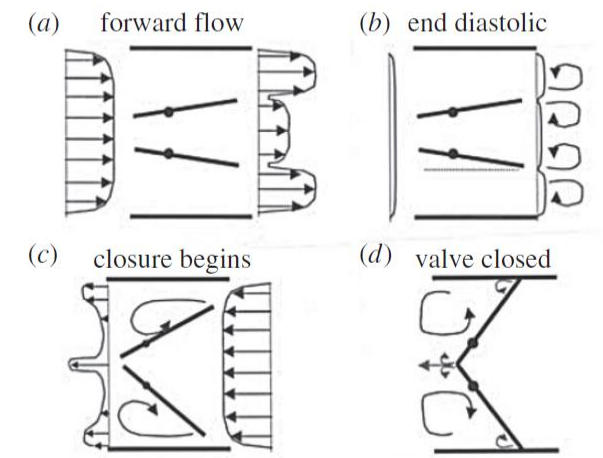
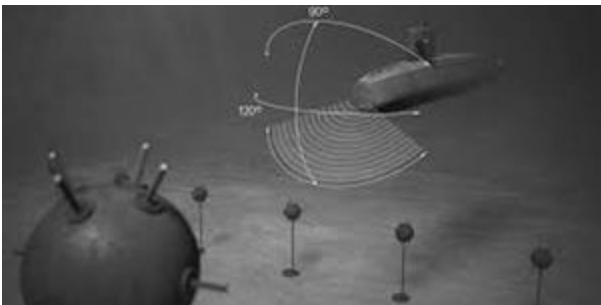
Cavitation research started with technical fluid mechanics in particular in marine technology (propellers, **sonar**, **underwater explosion**)

Cavitation limits performance of :

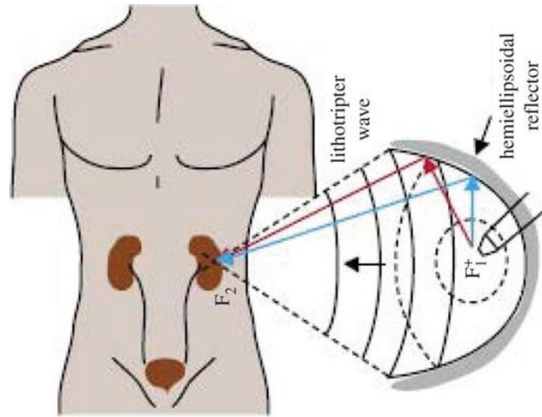
- ship propellers, blades
- pumps, motors

Cavitation enables:

- ultrasonic cleaning
- Laser ablation for drilling & hardening
- nano-particle production



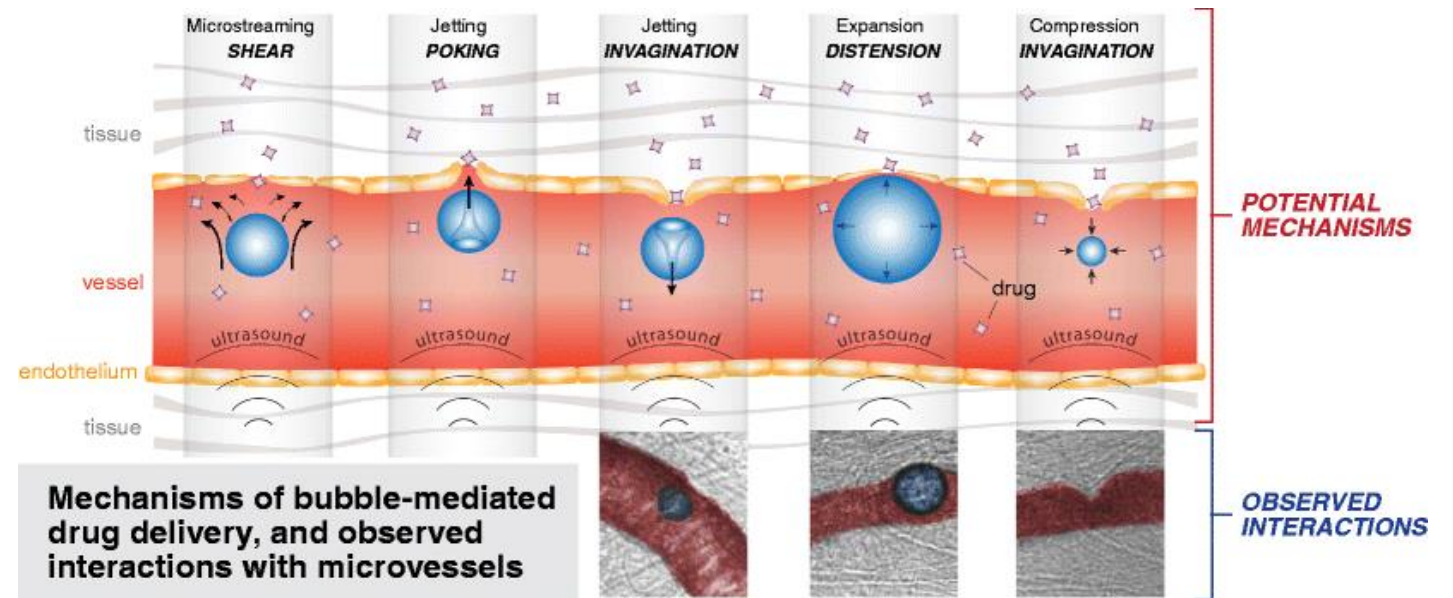
Cavitation in medicine and life science



Lithotripter / phacoemulsification / ultrasound fat cavitation

Different biomedical applications:

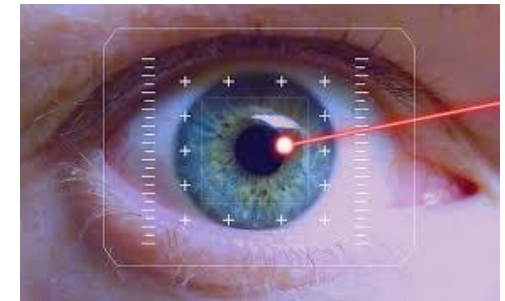
- Surgery / laser-tissue interaction
- drug delivery
- contrast enhancement in radiology



Cardiovascular applications of therapeutic ultrasound
Nazer et al., Journal of Interventional Cardiac Electrophysiology, 2014,

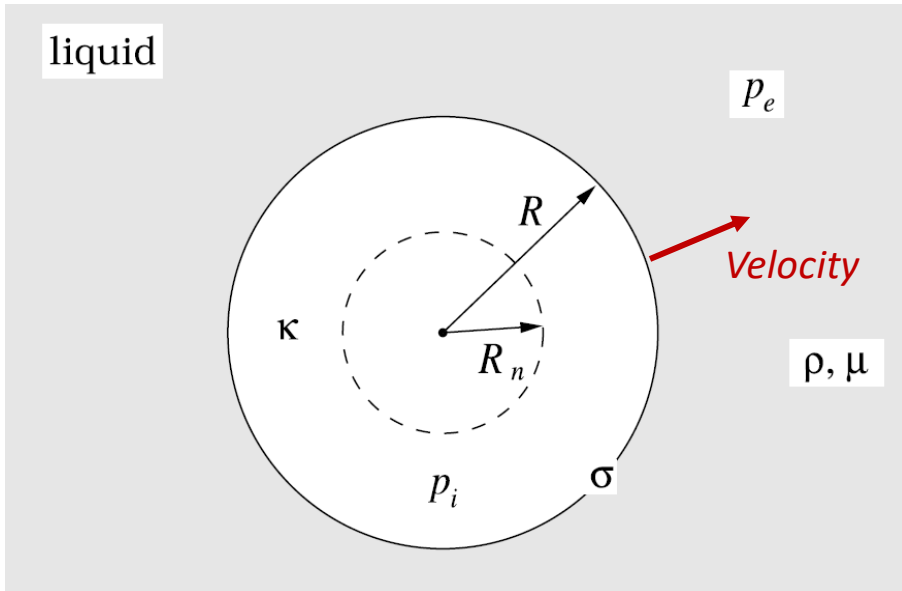
Surgery :

- Ophthalmology (e.g. lens membrane destruction following cataract surgery)
- Urology and gastroenterology (e.g., kidney and gall stone ablation and fragmentation)
- Cardiology and vascular surgery (e.g., laser ablation, removal of fibro-fatty, calcified arterial plaque)



Laser-induced cavitation!

Physics of bubbles and bubble oscillations



R: radius
 R_n : radius at equilibrium
 p_i : internal pressure
 p_e : external pressure
 σ : surface tension
 κ : adiabatic exponent
 ρ : density
 μ : (dynamics) viscosity

Interesting and extreme
 physical states and phenomena:

$$\rho R \ddot{R} + \frac{3}{2} \dot{R}^2 = p_i - p_e$$

Rayleigh equation (1917)

$$\rho R \ddot{R} + \frac{3}{2} \dot{R}^2 = p_{gn} \left(\frac{R_n}{R} \right)^{3\kappa} + p_v - p_{stat} - \frac{2\sigma}{R} - \frac{4\mu}{R} \dot{R} - p(t)$$

Rayleigh-Plesset equation

$$p - p_0 = B \left(\left(\frac{\rho}{\rho_0} \right)^m - 1 \right)$$

Tait eq.

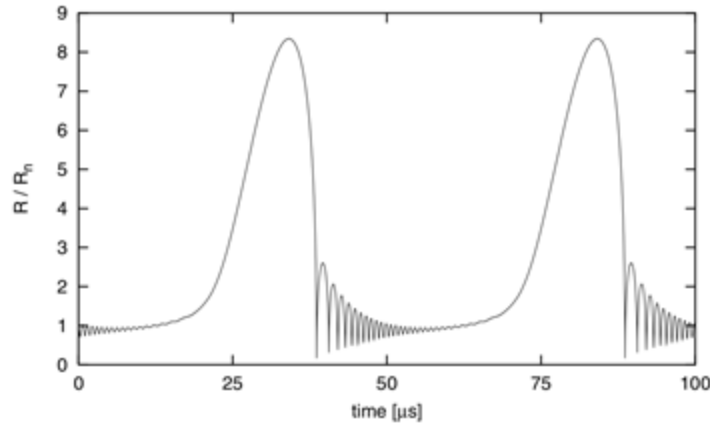
$$B = K/m \quad m = c_p/c_v \simeq 7$$

- bubble collapse: T ? p ? R_{min} ?
- *sonoluminescence*
- cavitation in artificial heart valves
- nature of the interfaces
- equation of state of H_2O : beyond Tait

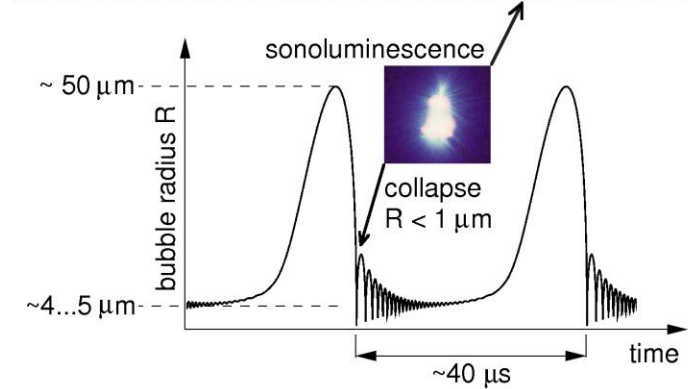
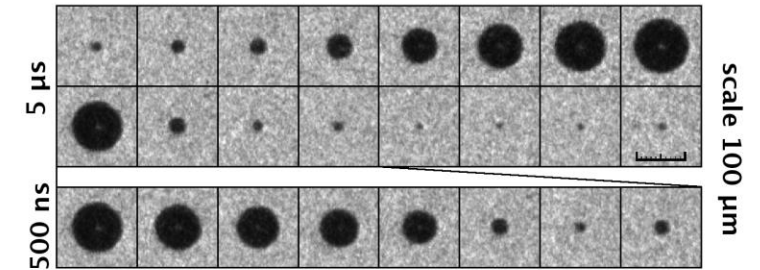
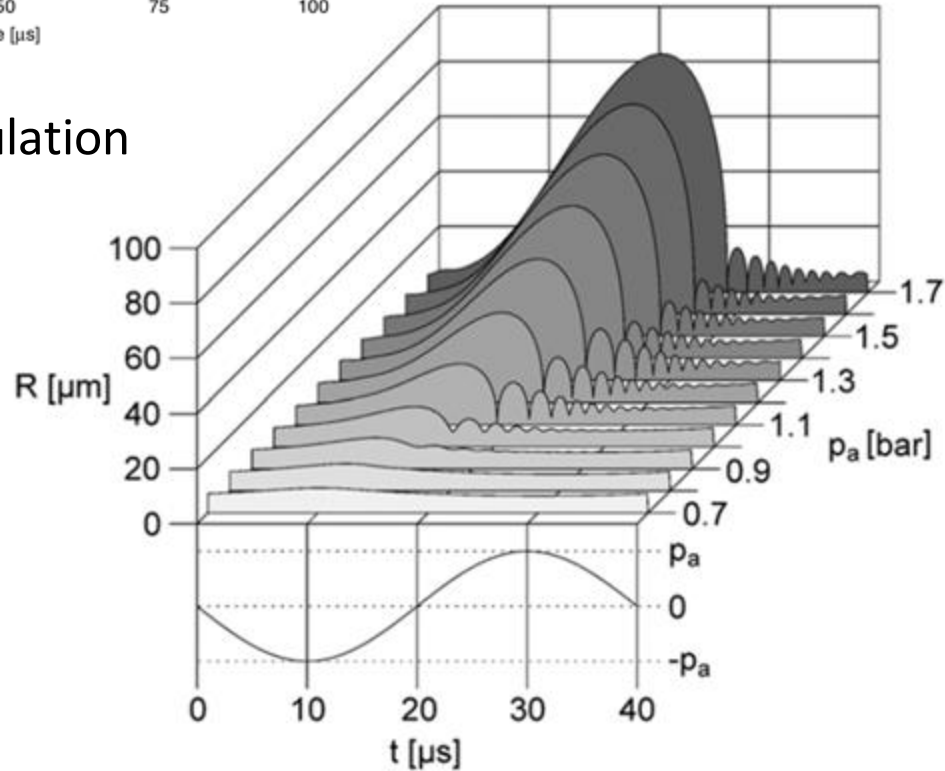
*...a lot is known, but all based on bubble trajectory,
 how about interior ? Always a clear phase separation ?*

Nonlinear dynamics, synchronized by acoustics

- suitable synchronized observation
- sub- μm radius @ collapse ?
- *how small ? density ? pressure ?*

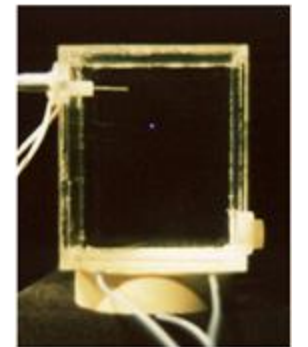


- numerical simulation

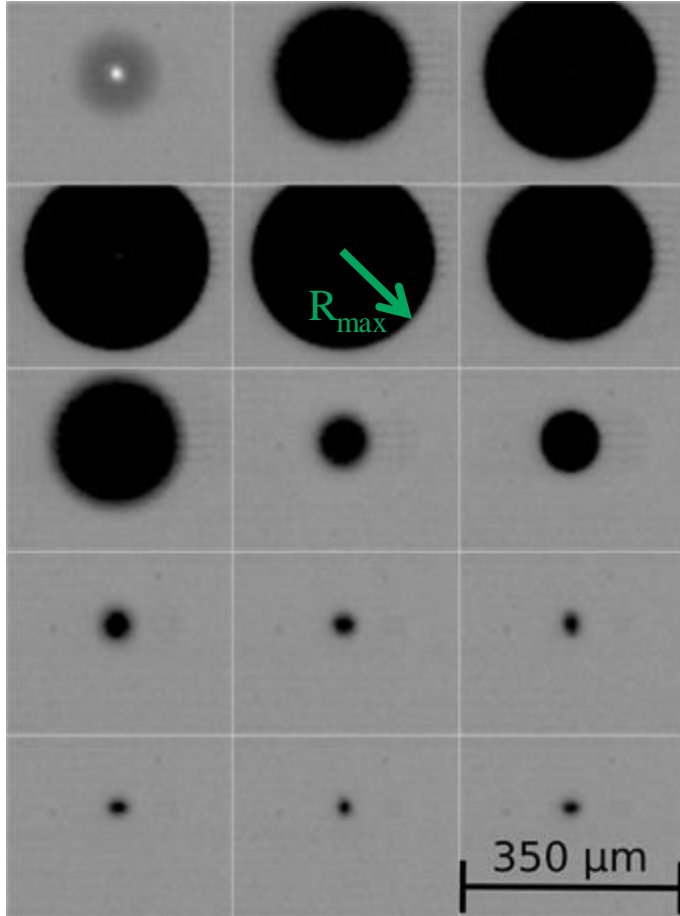


W. Lauterborn and T. Kurz,
Physics of bubble oscillations
Rep. Prog. Phys. 2010

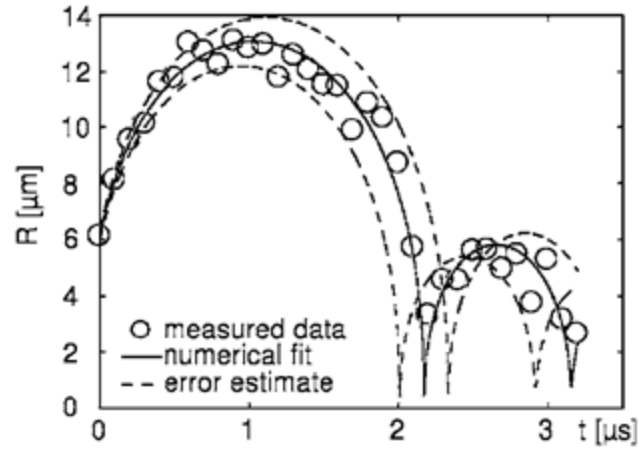
- sonoluminescence



Bubble seeding, energy & oscillations



300 kfps, 1μs shutter speed

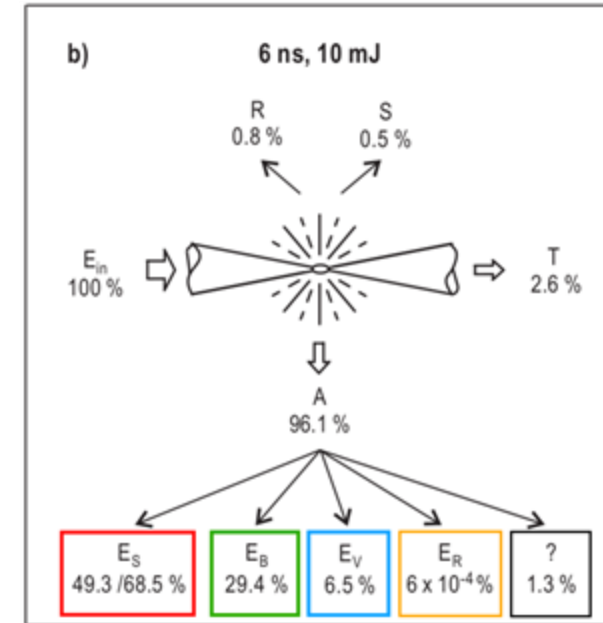


T. Kurz, D. Kröninger, R. Geisler, and W. Lauterborn, Phys. Rev. E (2006)

$$E_B = \frac{3\pi}{4} (p_0 - p_v) R_{max}^3$$

bubble energy determines maximum radius

- bubble nucleation
controlled by ps or *ns-laser pulse*)
- initial stages: plasma
- acoustic trapping
- bubble collapse



~59 % Shockwave energy

~30 % Bubble Expansion energy

~7 % Evaporation energy

~ $6 \cdot 10^{-4}\%$ Radiation energy

~1,3 % Dark Energy

Research questions

Plasma generated by
Optical breakdown
Multi-photon absorption
 $I > 10^{12} \text{ W/cm}^2$

A) For laser-generated cavitation bubble:

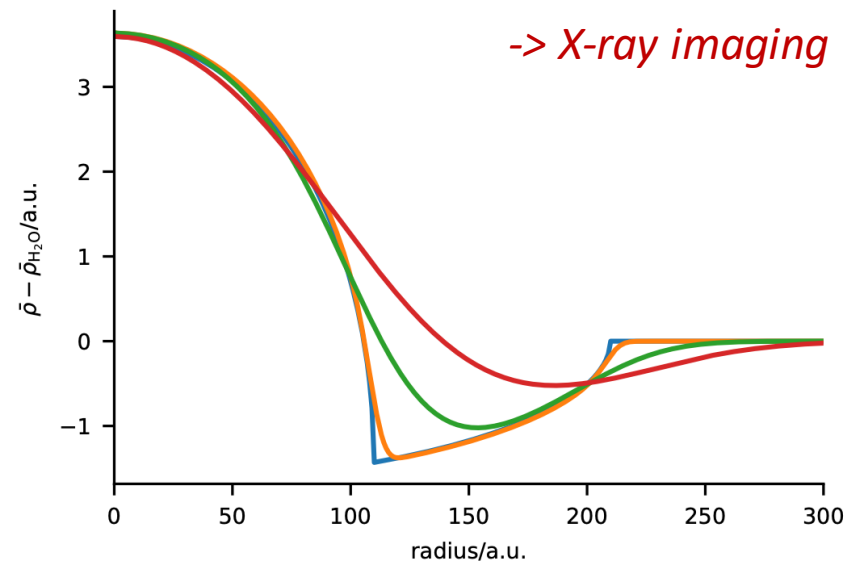
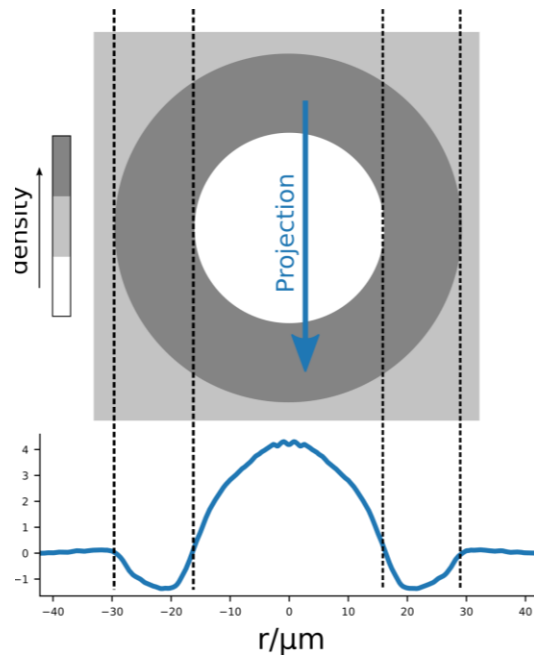
better understanding of **transition from plasma to cavitation bubble**

B) Shock wave effects can be useful or unwanted depending on applications

-> detailed knowledge of the shock emission processes and the **properties of the shock wave** required

C) Fundamental physics: extreme conditions at **bubble collapse** (sonoluminescence, $T > 10^4 \text{ K}$)

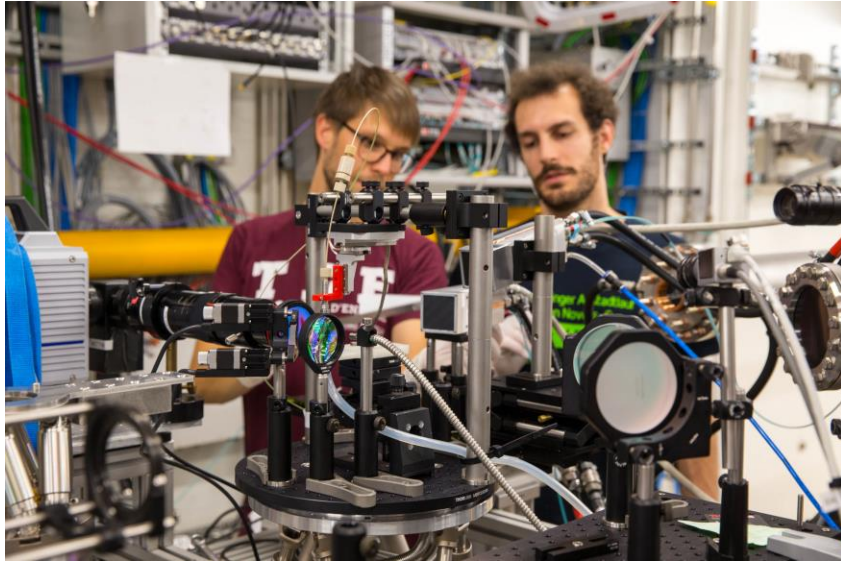
*Challenge: Spatio-temporal resolution, contrast
refraction, scattering, opacity, resolution –*



- bubble evolution
- density of the shockwave ?
- sharp interfaces gas/water ?

radial density profile !

email from XFEL (2.11.18) – a disruptive event

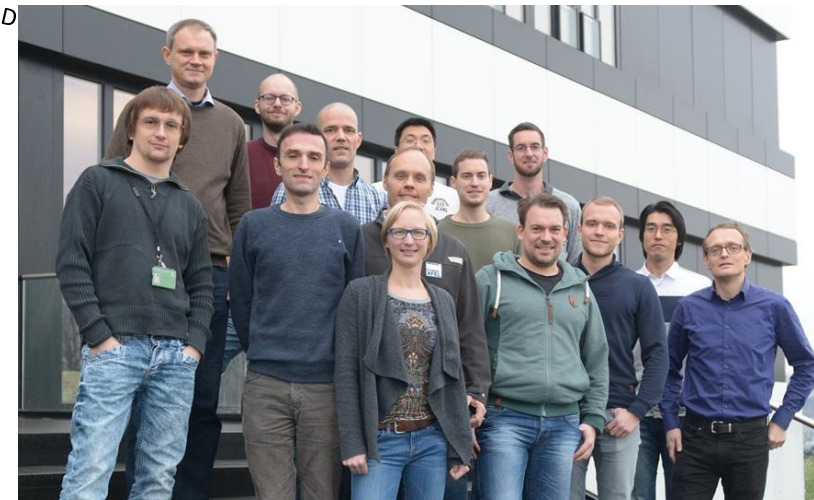


You cannot count on XFEL beamtime
For your Ph.D. or master project

... so when it comes, you have to change plans...

Am 02.11.18 um 14:42 schrieb European XFEL User Office:
> Main Proposer: Dr. Johannes Hagemann / email:
> johannes.hagemann@desy.de
>
> Principal Investigator: Prof. Dr. Tim Salditt / email: tsaldit@gwdg.de
>
> *Your Proposal No. 2207 at the European XFEL, allocation cycle 201802*
>
> *Title: Cavitation Dynamics Studied by Time-Resolved High-Resolution
> X-Ray Holography*
> **
>
> Dear Main Proposer and Principal Investigator for proposal No. 2207,
>
> Many thanks for submitting the above proposal in the 3rd call for
> early user experiments at the European XFEL. It is our pleasure to
> confirm that the project has been allocated beamtime at the MID
> follows:
>
> *Start date: 05 June 2019 (Day shift) *
>
> *End date: 09 June 2019 (end of last shift) *
>
> *Total shift allocated: 5*

...disruptive for everybody!



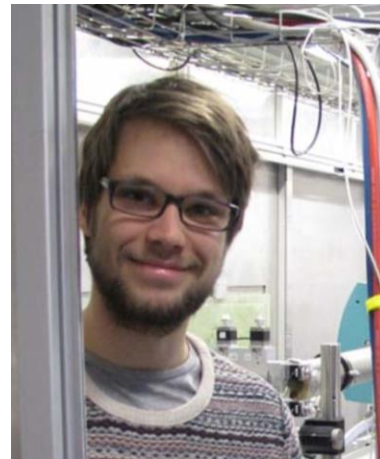
large collaboration & strong individual contributions

Malte Vassholz, Markus Osterhoff, Hannes Hoeppe, Juan Rosello, Robert Mettin, Thomas Kurz, Tim Salditt
Universität Göttingen, III-Phys. Inst.-Biophysik & Institut für Röntgenphysik

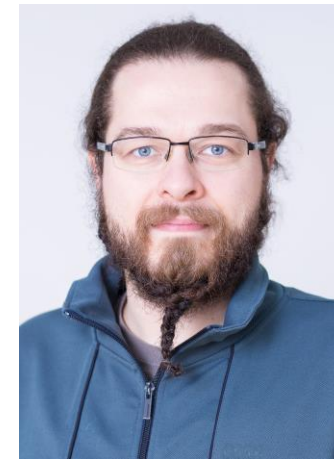
Johannes Hagemann, Frank Seiboth, Andreas Schropp, Christian G. Schroer,
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MID / XFEL

- UAC MID 06/2019 @14keV
- Beamtime 10/ 2019 @17.8keV
(p2207 & p2545 Hagemann/Salditt)



Malte Vassholz



Johannes Hagemann



Markus Osterhoff

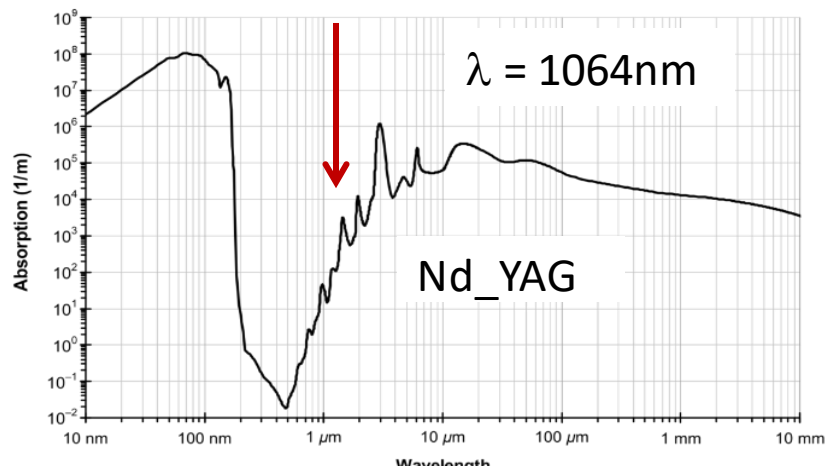
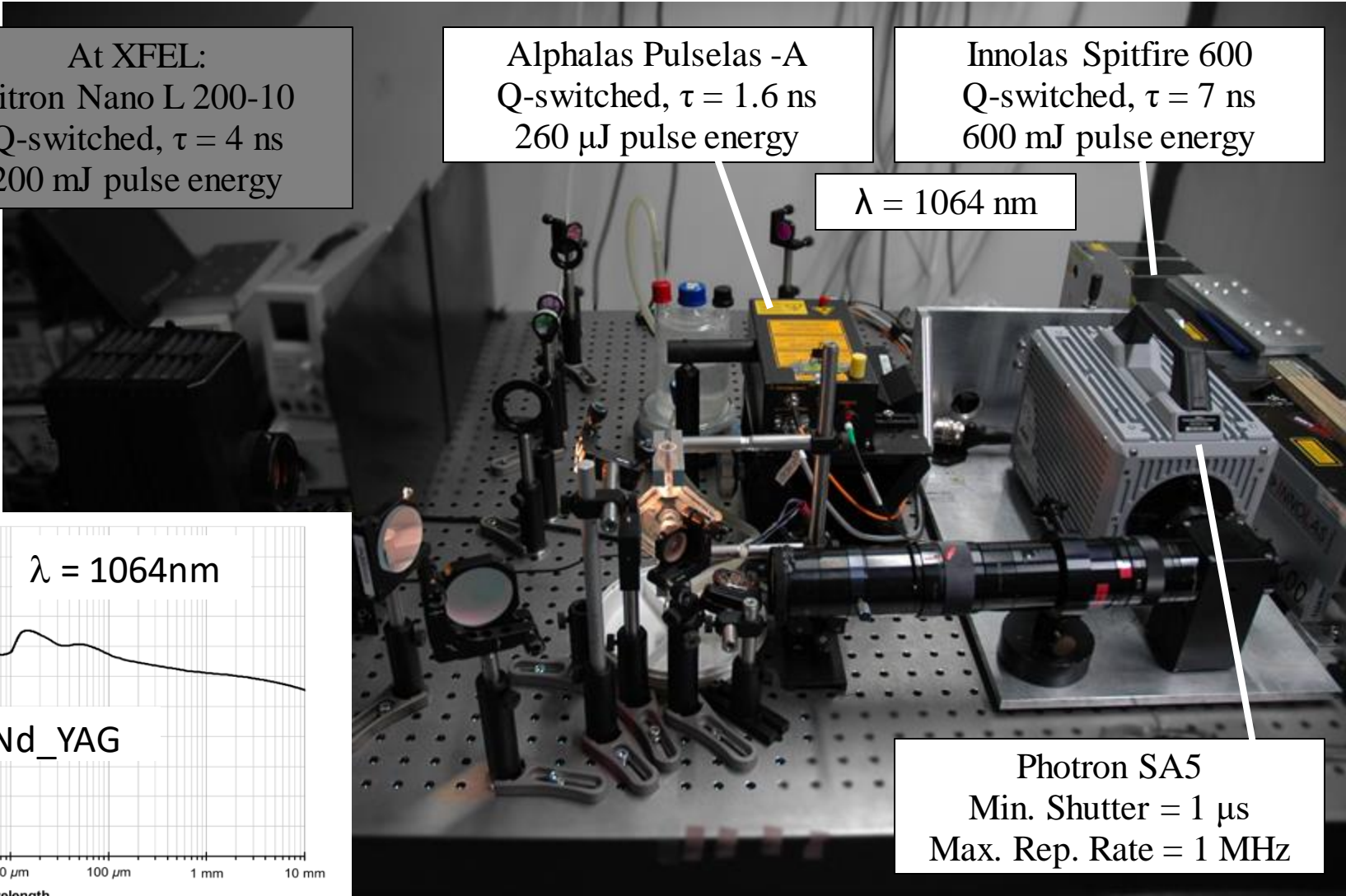
High speed optical imaging

At XFEL:
Litron Nano L 200-10
Q-switched, $\tau = 4$ ns
200 mJ pulse energy

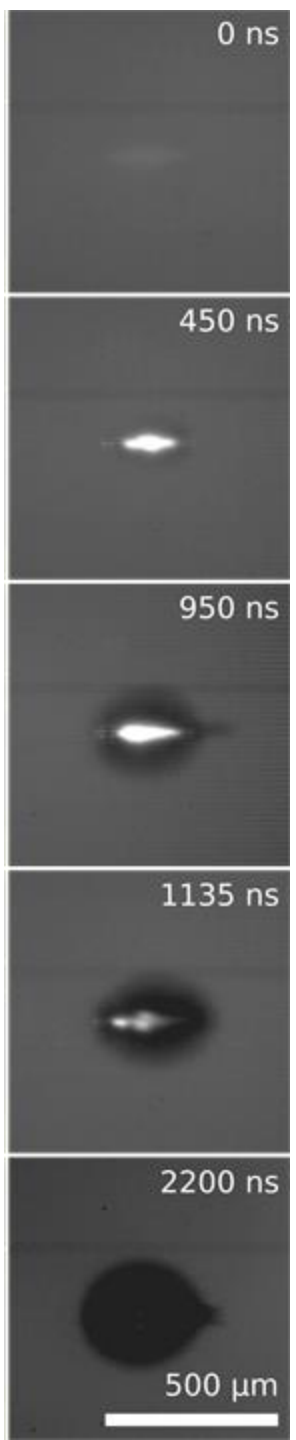
Alphas Pulselas -A
Q-switched, $\tau = 1.6$ ns
260 μ J pulse energy

Innolas Spitfire 600
Q-switched, $\tau = 7$ ns
600 mJ pulse energy

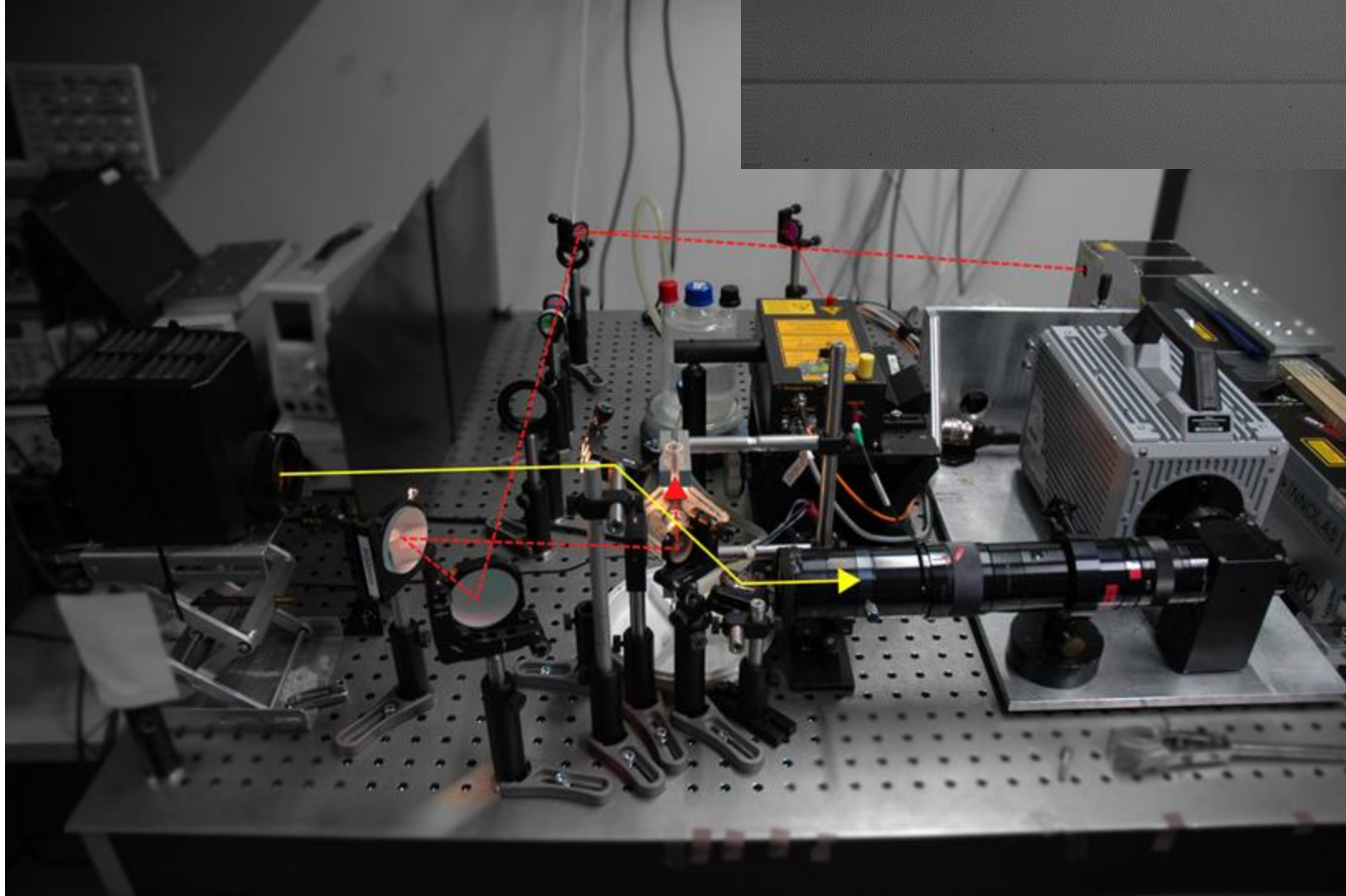
$\lambda = 1064$ nm



Photron SA5
Min. Shutter = 1 μ s
Max. Rep. Rate = 1 MHz

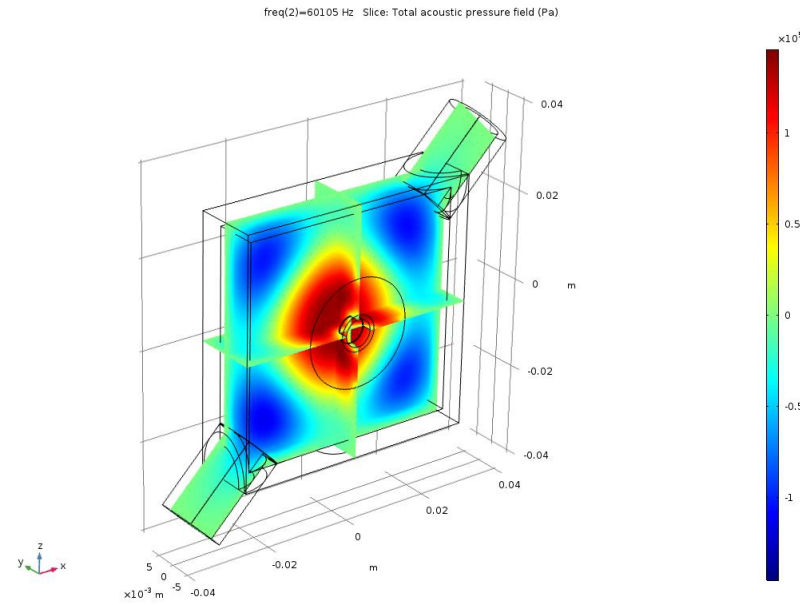
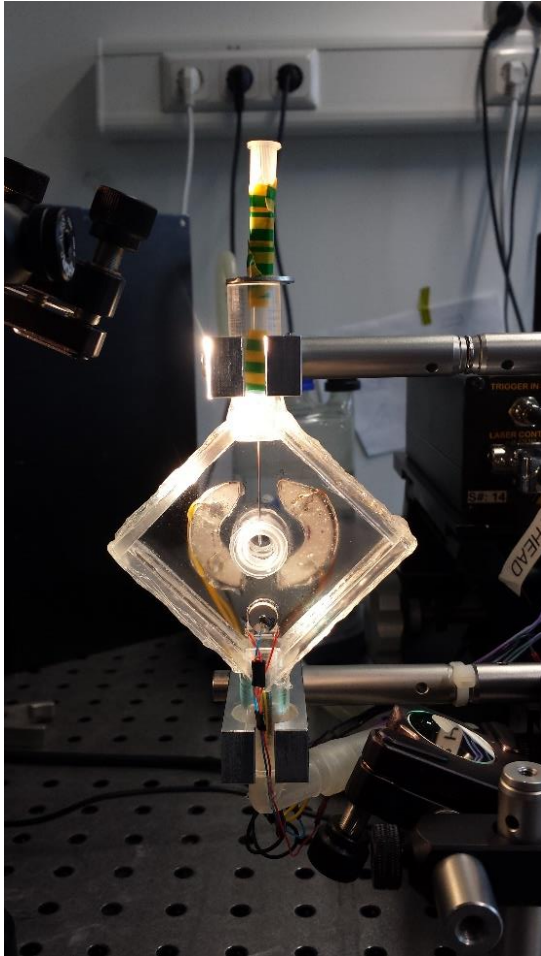


High speed optical imaging



Juan Rosello, Hannes Hoeppe

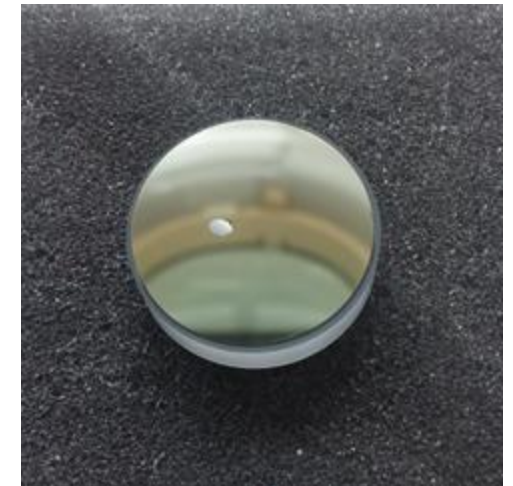
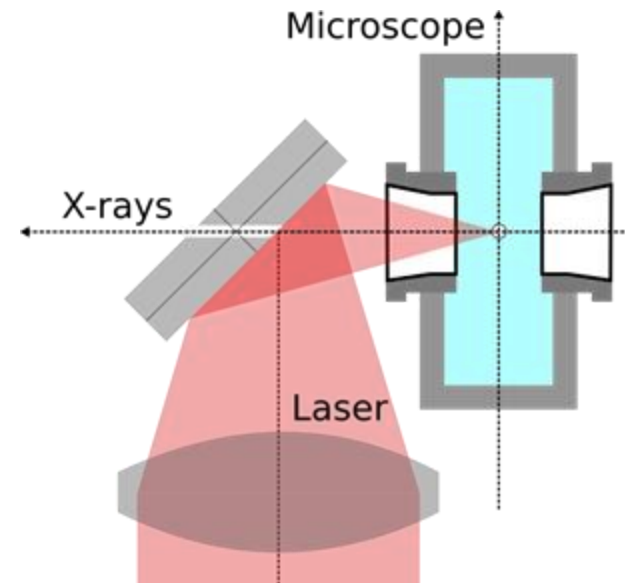
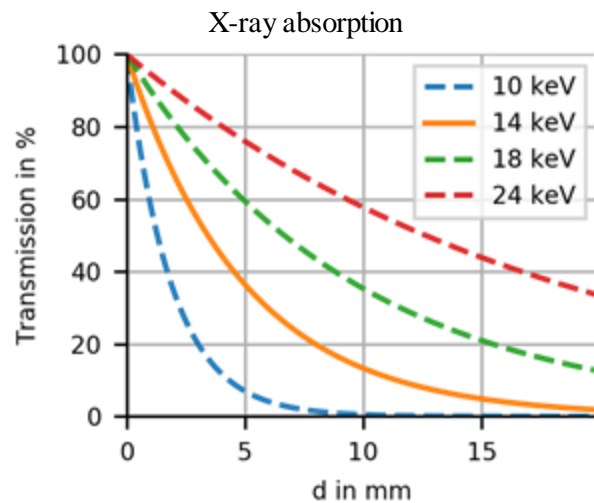
Cavitation cuvette x-ray compatible...



- suitable for acoustic driving /trapping
- **Low x-ray absorption vs. Stability**
- compatible with XFEL defocused beam
- compatible with IR laser
- compatible speed optical camera

-> anti-parallel XFEL and IR-beams

Mirror with drilled hole



Malte Vassholz, Hannes Hoeppe

High-speed optical camera

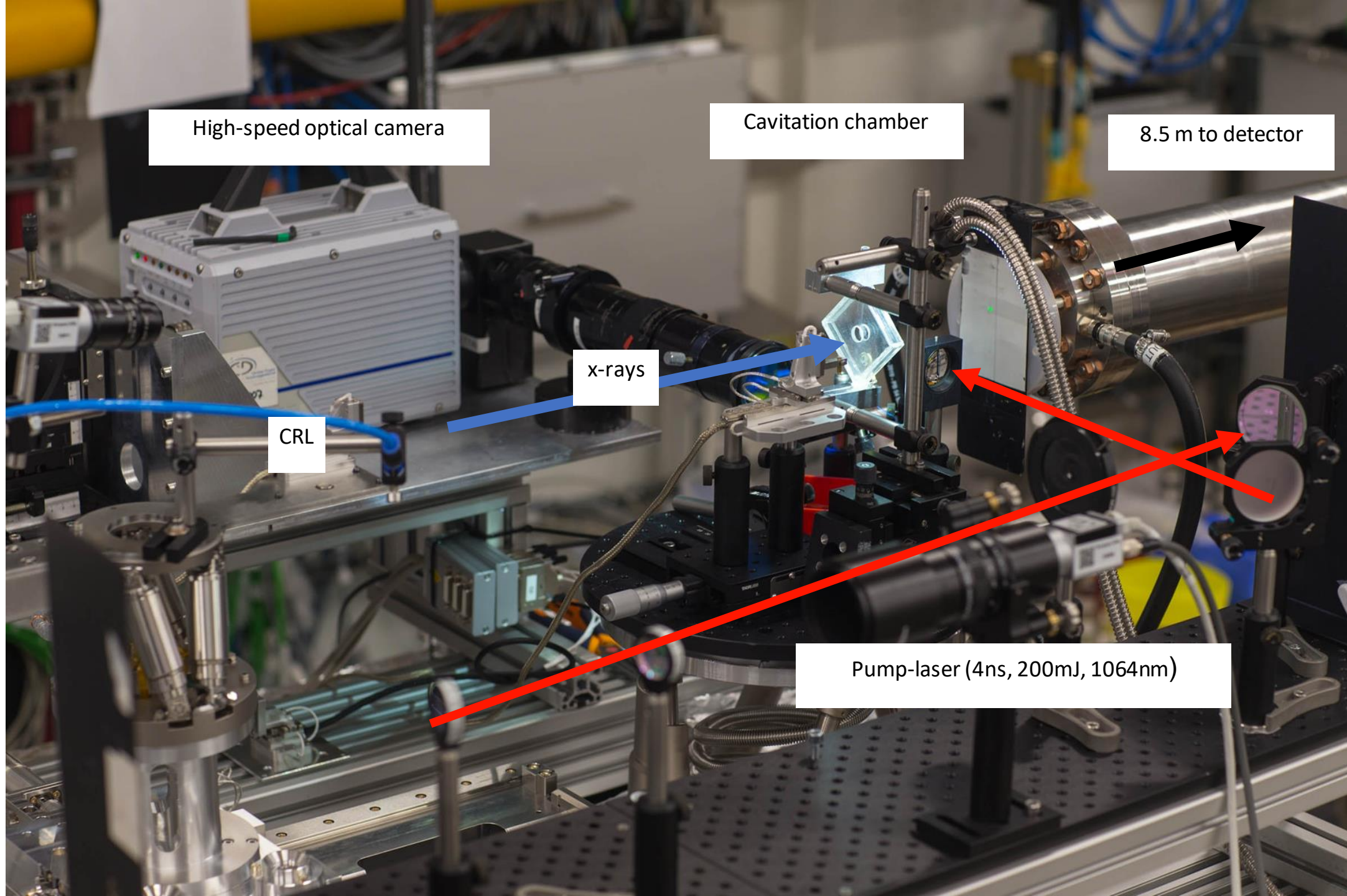
Cavitation chamber

8.5 m to detector

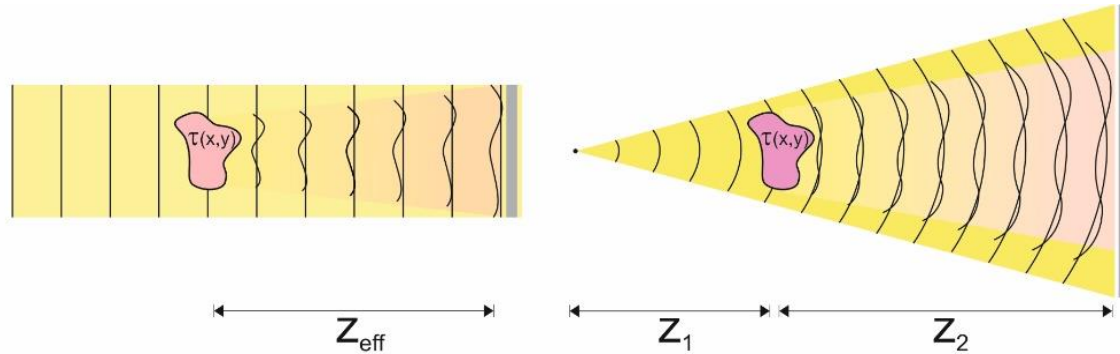
x-rays

CRL

Pump-laser (4ns, 200mJ, 1064nm)

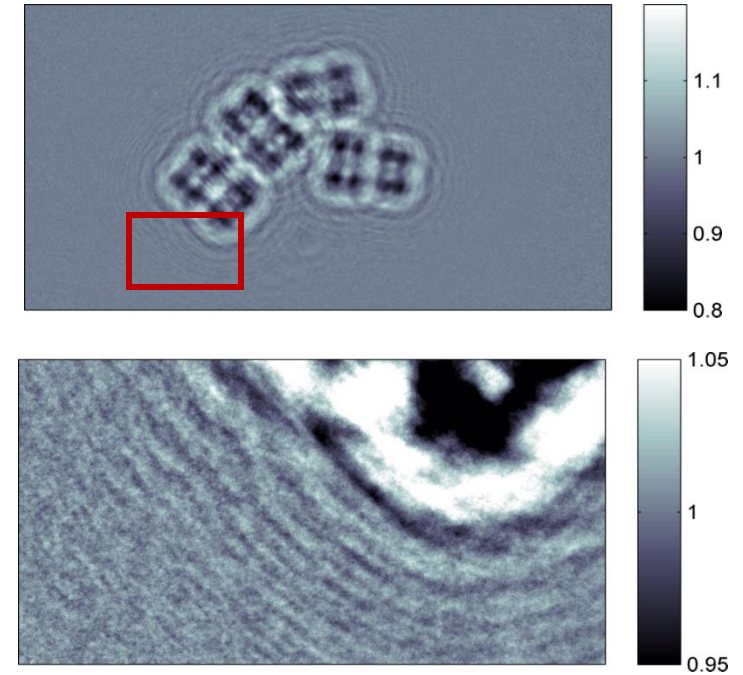


Full-field X-ray imaging / (inline) holography



$$M = \frac{z_1 + z_2}{z_1}$$

$$z_{eff} = \frac{z_1 z_2}{z_1 + z_2}$$

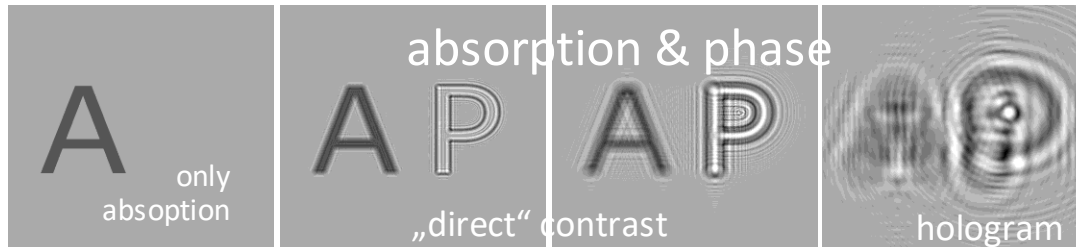


object with complex transmission function $\tau(x,y)$

$$\psi_z = FT^{-1} [\exp[iz\sqrt{k^2 - k_x^2 - k_y^2}] FT[\psi_0]]$$



Absorption Phase



Fresnel number F

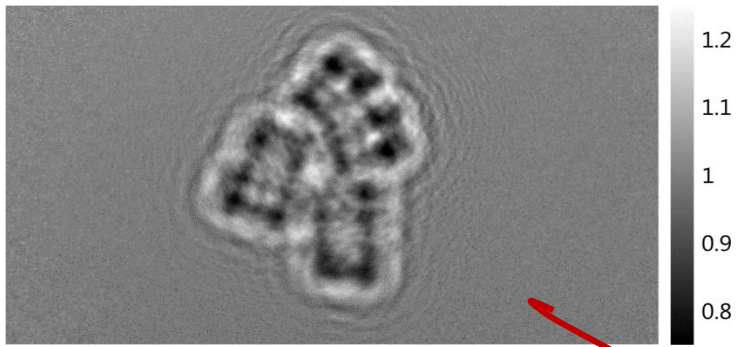
SR: Blurring of fringes !
Low signal in water

XFEL:

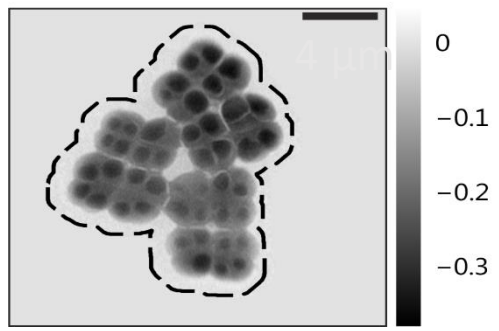
- **no motional blurring,**
- **ultimately sharp holograms !**
- **outrun radiation damage**

Holographic phase retrieval

- high numerical aperture / high resolution
- quantitative reconstruction beyond linearisation
- probe aberrations (wavefront, coherence)

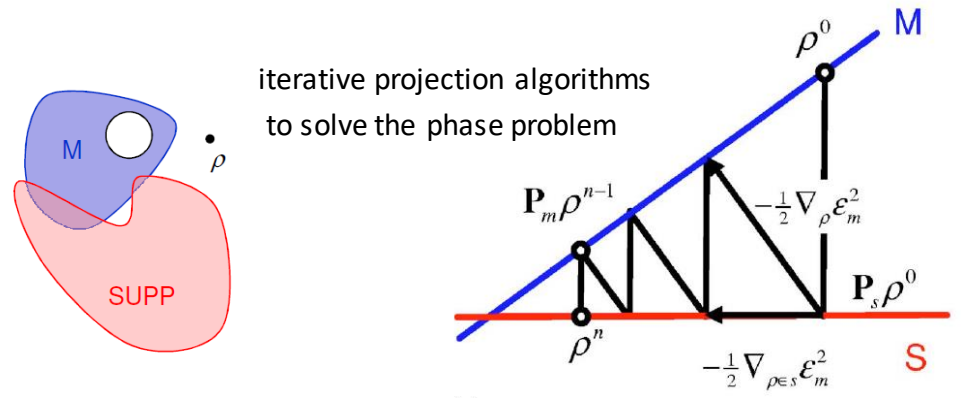


bacterial cells (Deinococcus radiodurans)



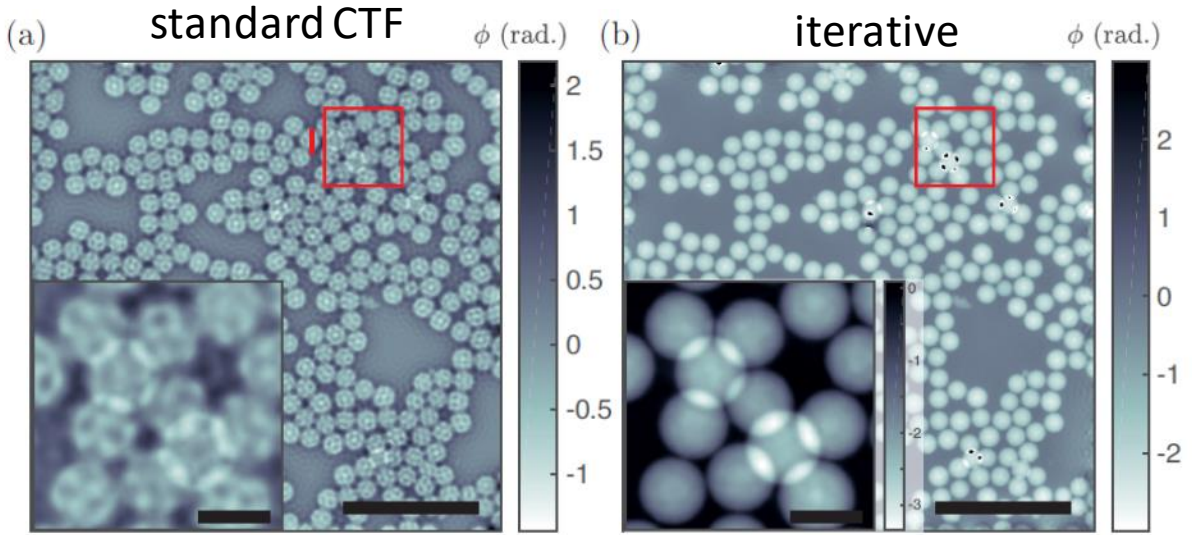
key challenge:
phase retrieval!

Bartels, Krenkel, Haber, Salditt Phys.Rev.Lett. 2015



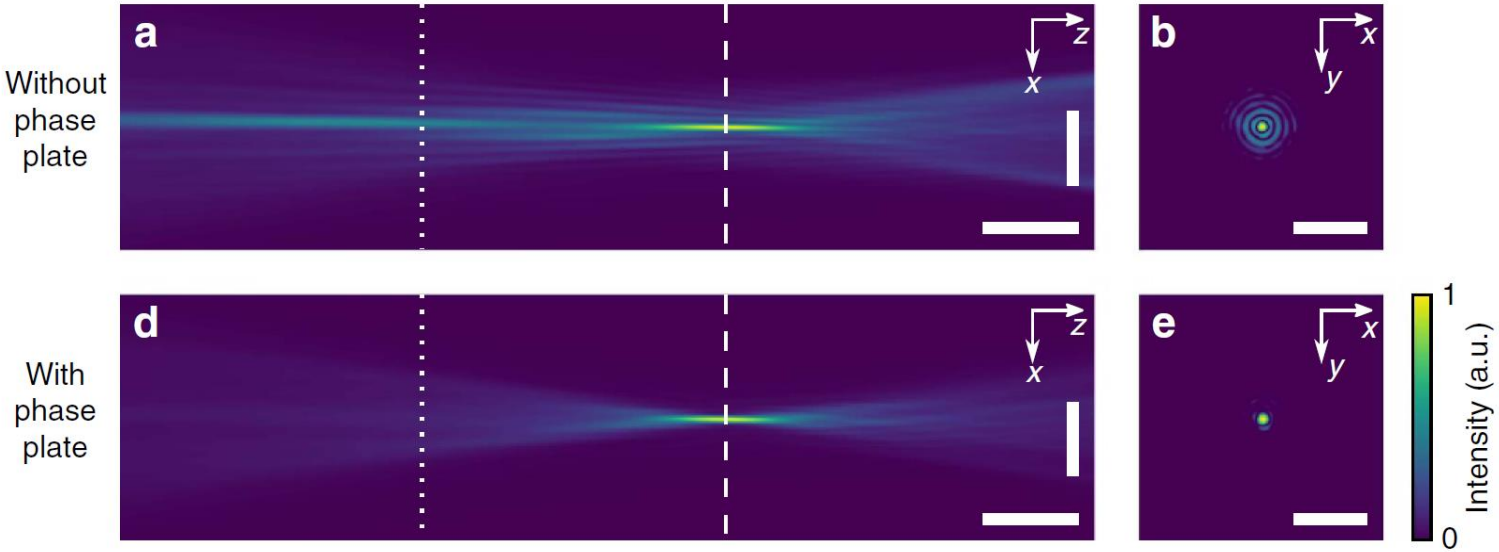
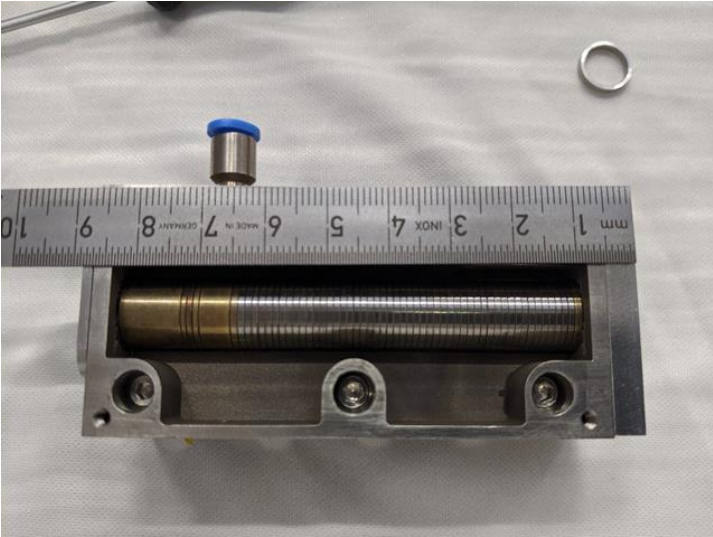
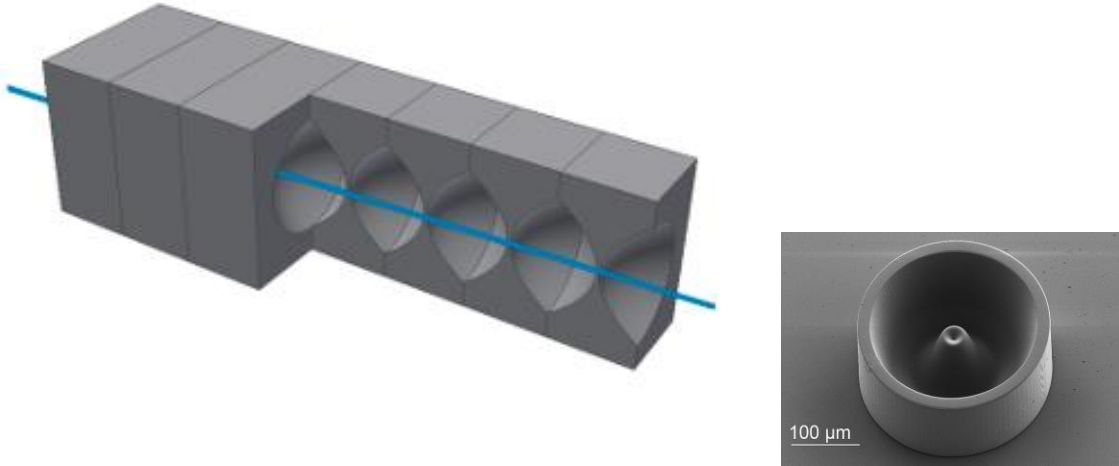
adaptation for near-field:

Giewekemeyer et al, PRA 2011; Hagemann et al. 2015, 2016; Robisch et al., 2015; Krenkel et al. Acta Cryst 2017



Hagemann, Töpperwien, Salditt Appl.Phys.Lett. 2018

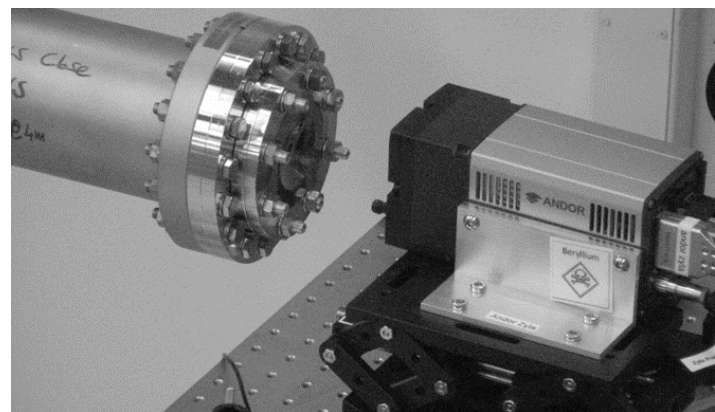
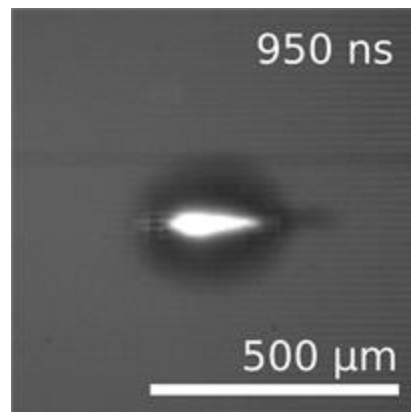
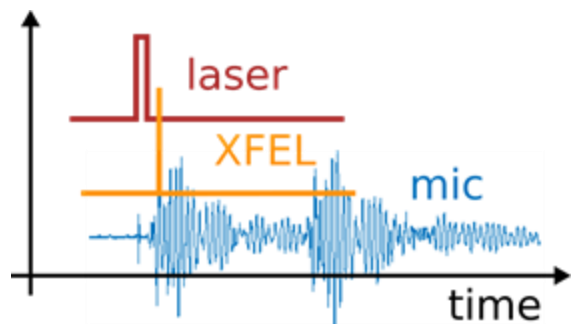
CRL (Be) focusing to $\sim 100\text{nm}$



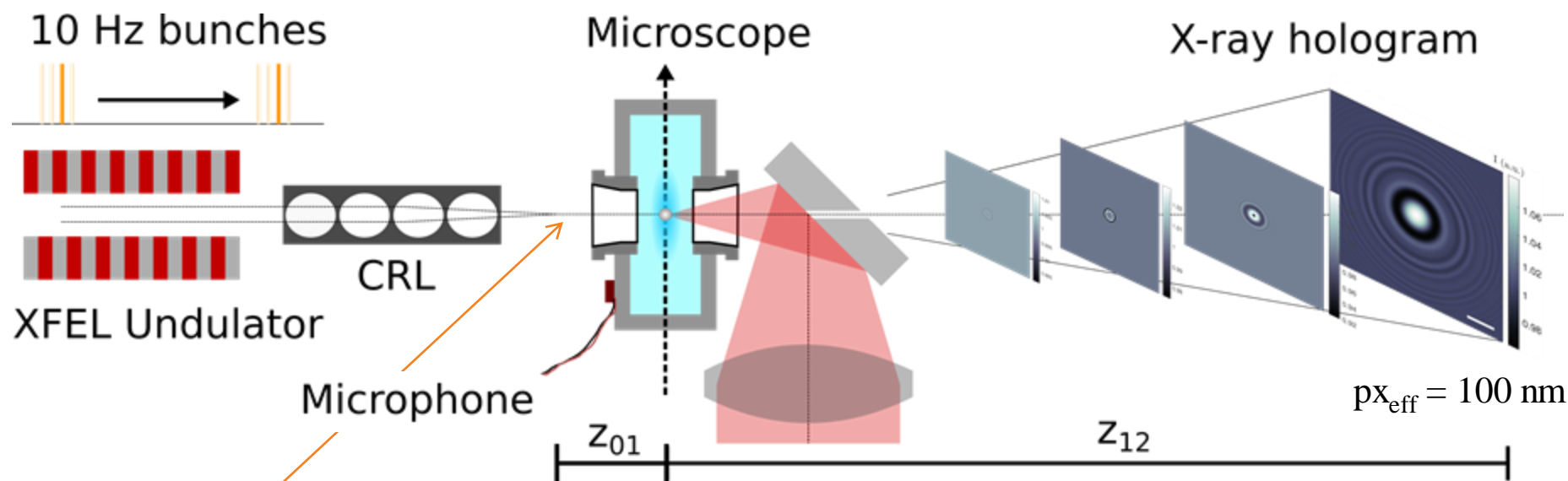
N=50 Be CRL
R=50 μm
D=300 μm
f=289mm
 $\Delta \sim 100\text{nm}$

Seiboth et al., Nat. Comm. (2017)

Settings at MID / exp. parameters



Andor Zyla 5.5 HF
 20μm LuAG:Ce
 px=6.5μm
 z=8.859m



10 Hz
 600 μJ / pulse
 $3 \cdot 10^{11}$ ph / pulse
 $\tau < 100\text{fs}$
 E = 14keV (UAC)
 E = 18keV (Oct.19)

$W_{0,x} = 100 \text{ nm}, \text{div} = 0.6 \text{ mrad}$

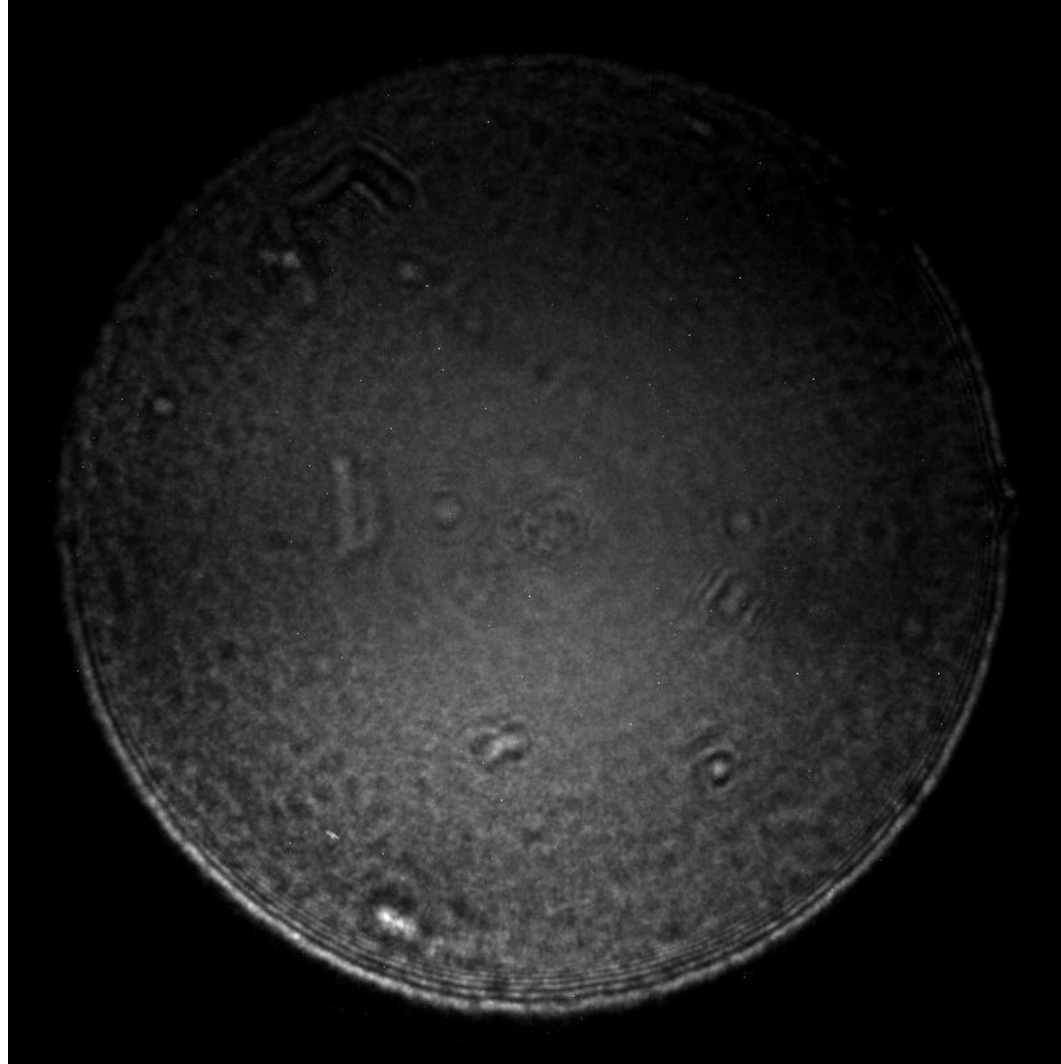
$M = 61 \quad px_{\text{eff}} = 107 \text{ nm}$

And this is how it looks – *meet your probe* !

in terms of

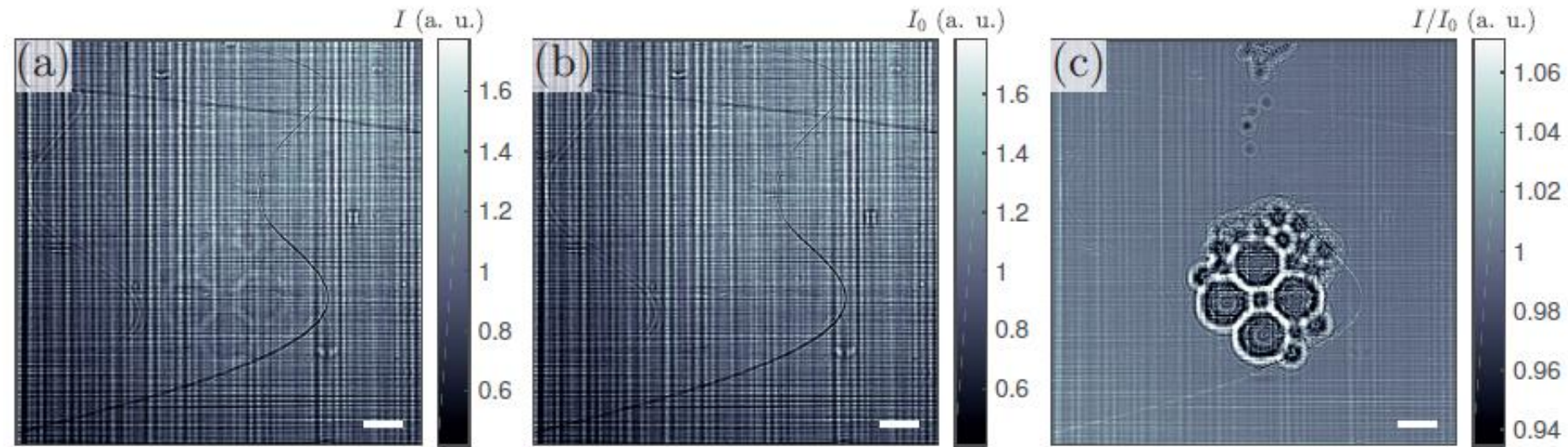
- *divergence,*
- *pointing stability*
- *signal*

*it is all actually
not too bad
for a single pulse!*



*But how to
perform the
flat field correction?*

The empty beam problem in holography



Simultaneous probe and object reconstruction for the near-field
 A.L. Robisch, K. Kröger, A. Rack, T.Salditt, N.J.Phys. 2015

Reconstruction of wave front and object for inline holography
 from a set of detection planes
 J.Hagemann, A.L.Robisch et al., Optics express 2014

Validity of the empty-beam correction in near-field imaging
 C. Homann, T.Hohage, J. Hagemann, A.L. Robisch, T.Salditt
 Physical Review A 2014

Holographic imaging with a hard x-ray nanoprobe: Ptychographic versus conventional phase retrieval
 A.-L. Robisch et al., J. Wallentin, A. Pacureanu, P. Cloetens, and T. Salditt Opt.Lett. 2016

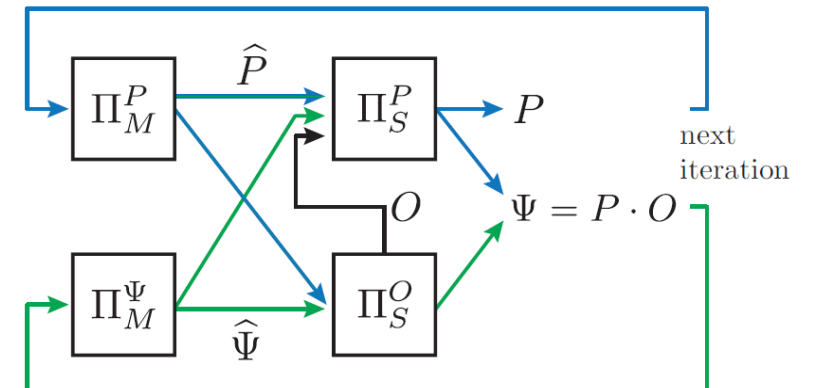
**flat field correction
 is flawed!**



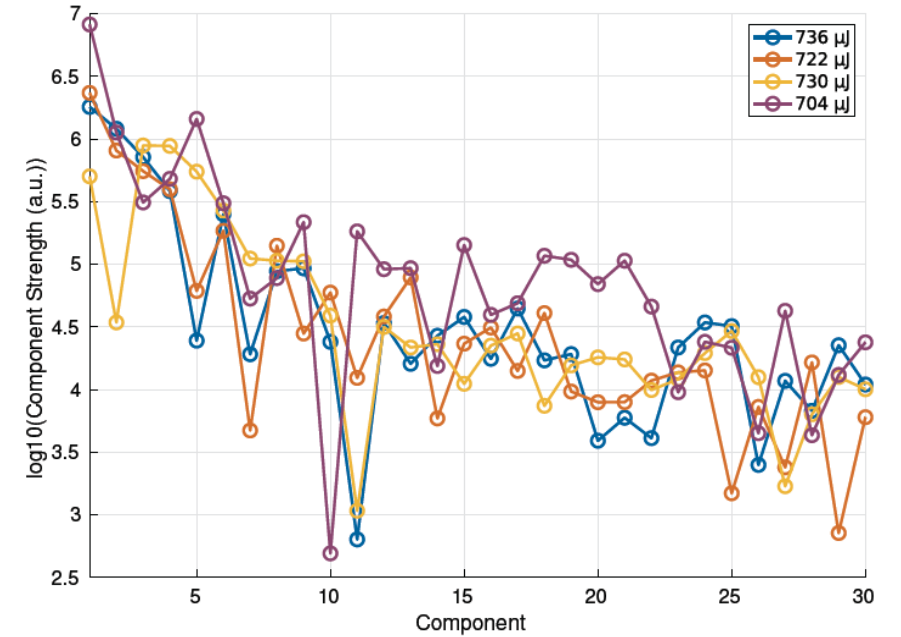
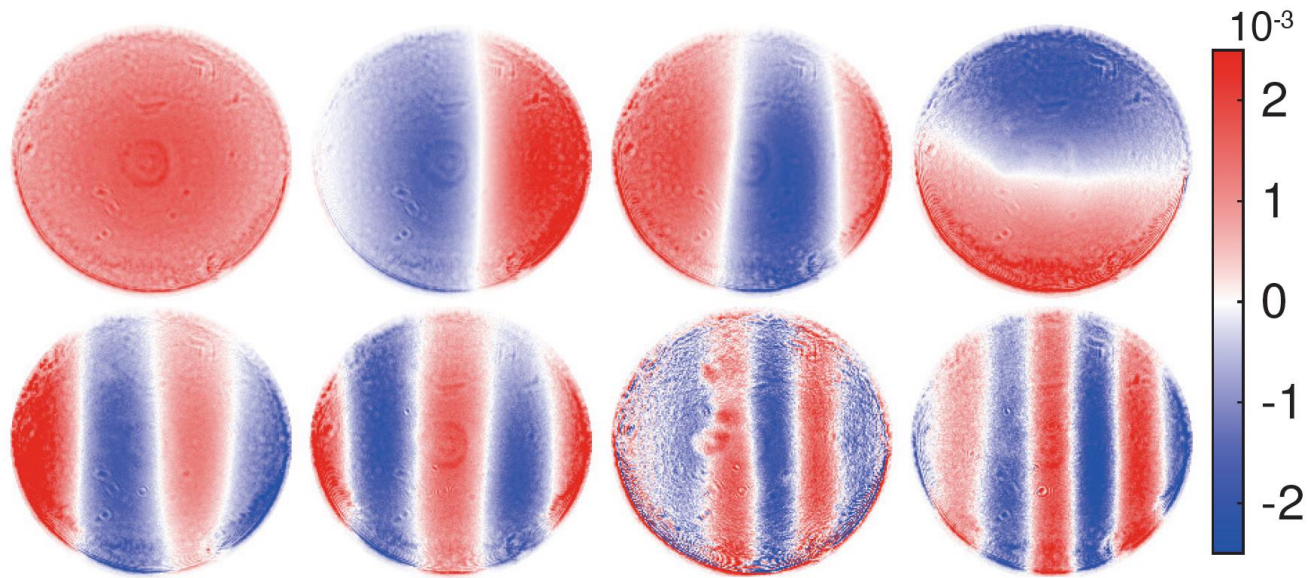
**Divide and update: towards single-shot object
 and probe retrieval for near-field holography**

JOHANNES HAGEMANN^{1,2} AND TIM SALDITT^{1,3}

¹Universität Göttingen, Institut für Röntgenphysik, Friedrich-Hund-Platz 1, 37077 Göttingen Germany



Let's decompose (PCA analysis)



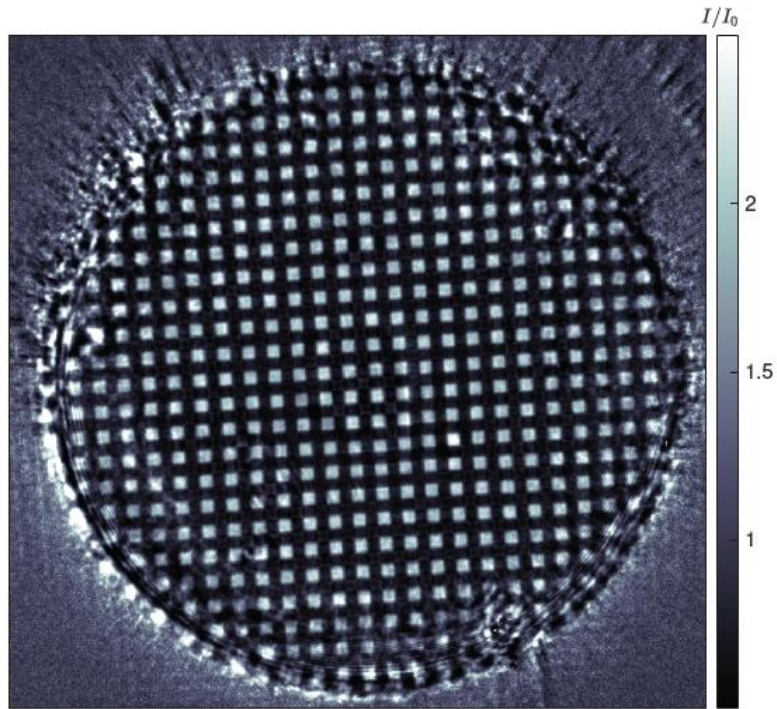
Based on:

Dynamic intensity normalization using eigen flatfields in X-ray imaging
V. V. Nieuwenhove et al. , Optics Express 23(21), 27975 (2015)

Johannes Hagemann

Let's decompose (PCA analysis)

now imaging works!

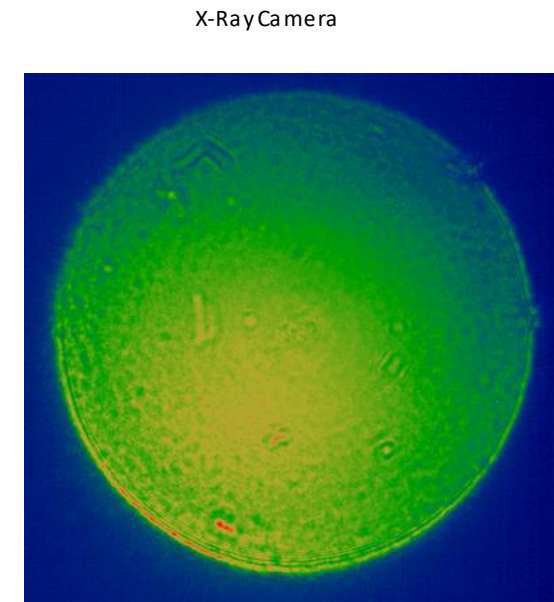
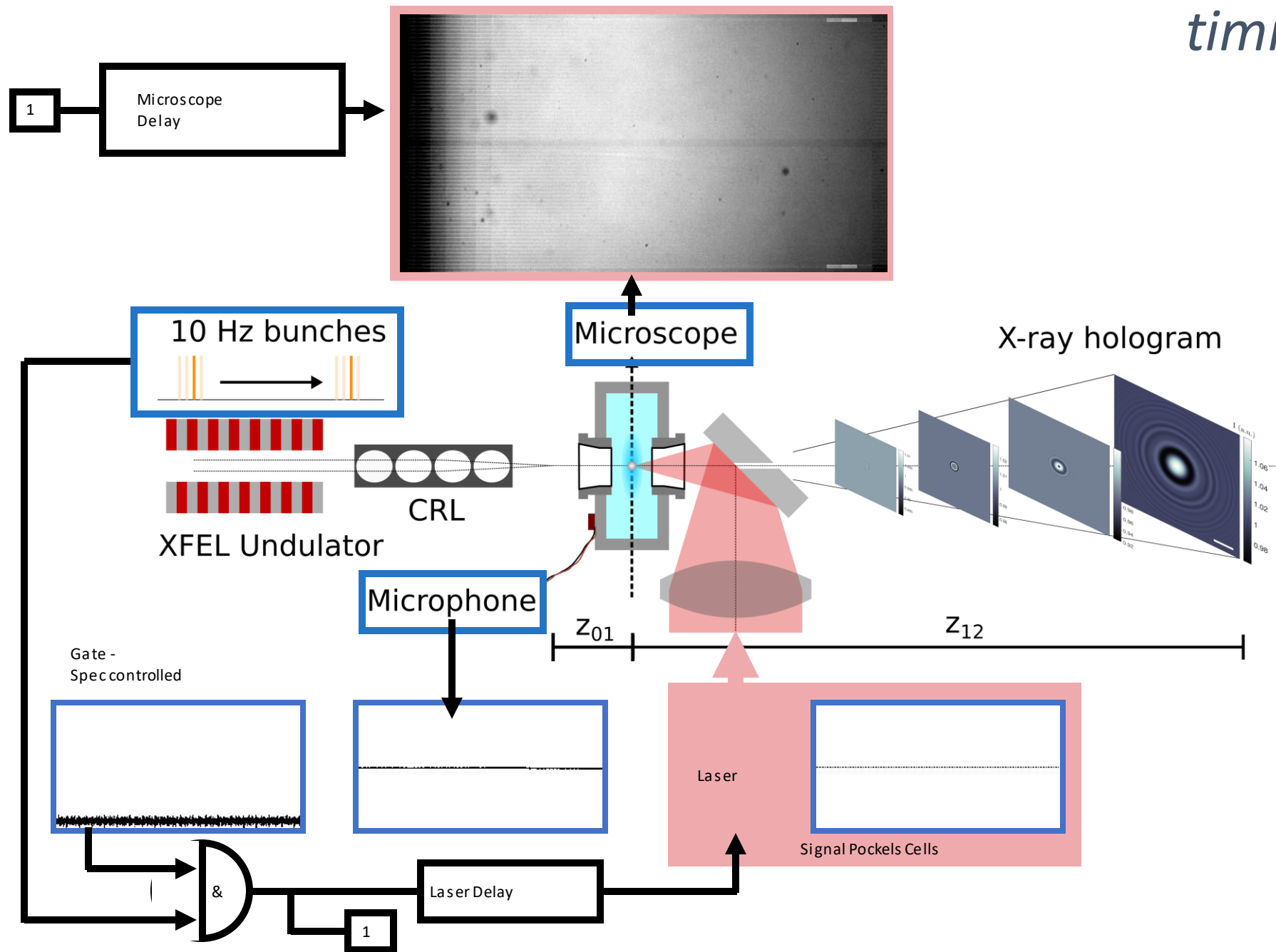


6 μ mAu mesh



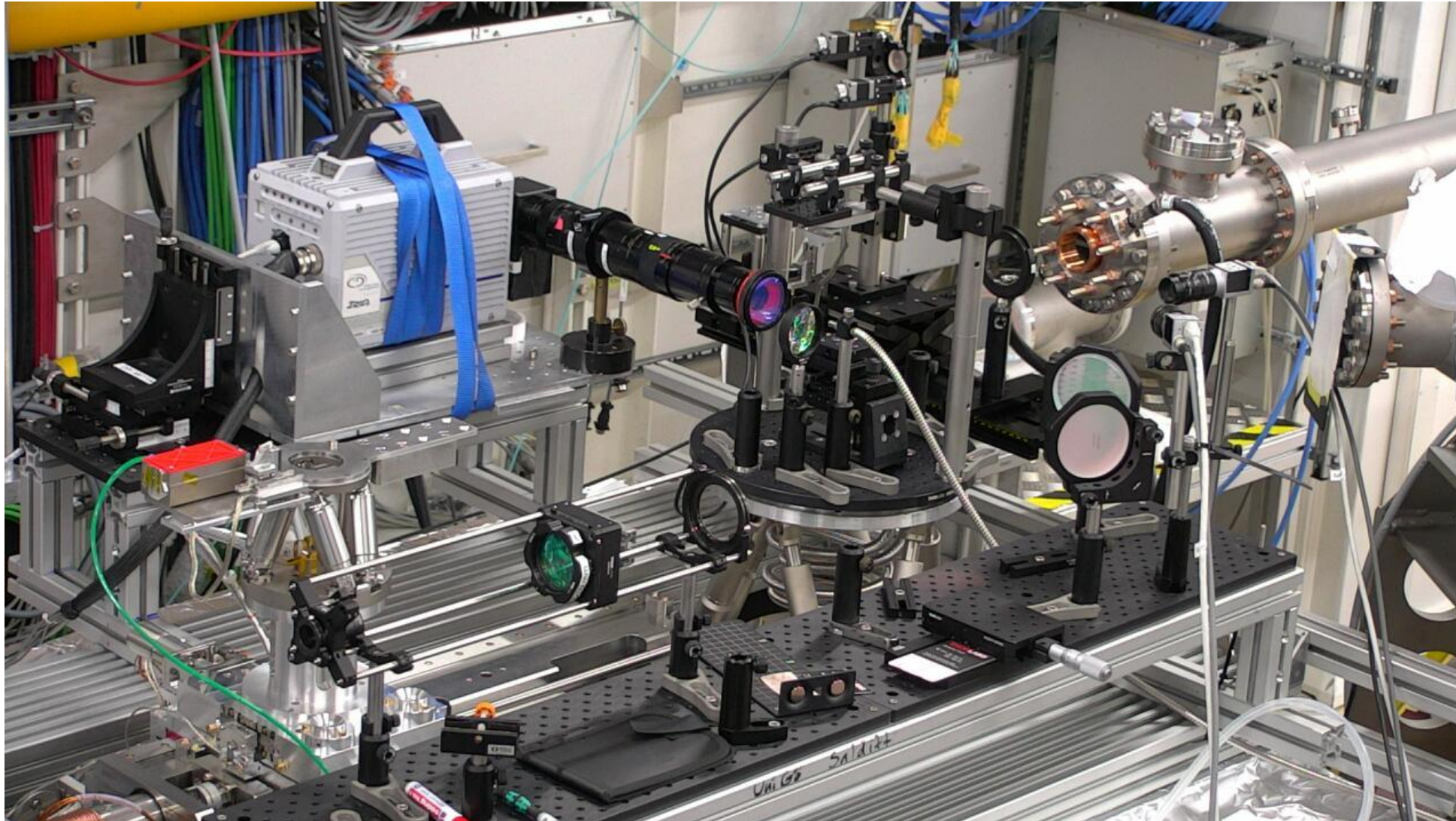
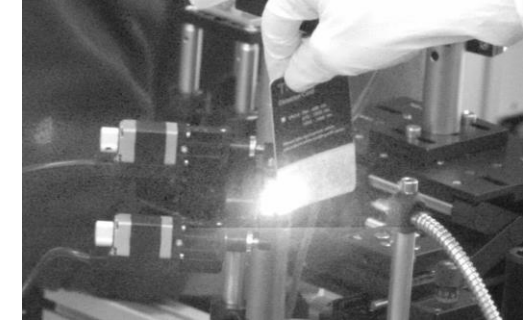
laminar water jet

timing & synchronisation



scheme works nicely – but we miss the bubble

*A good timing scheme
is not sufficient ...*

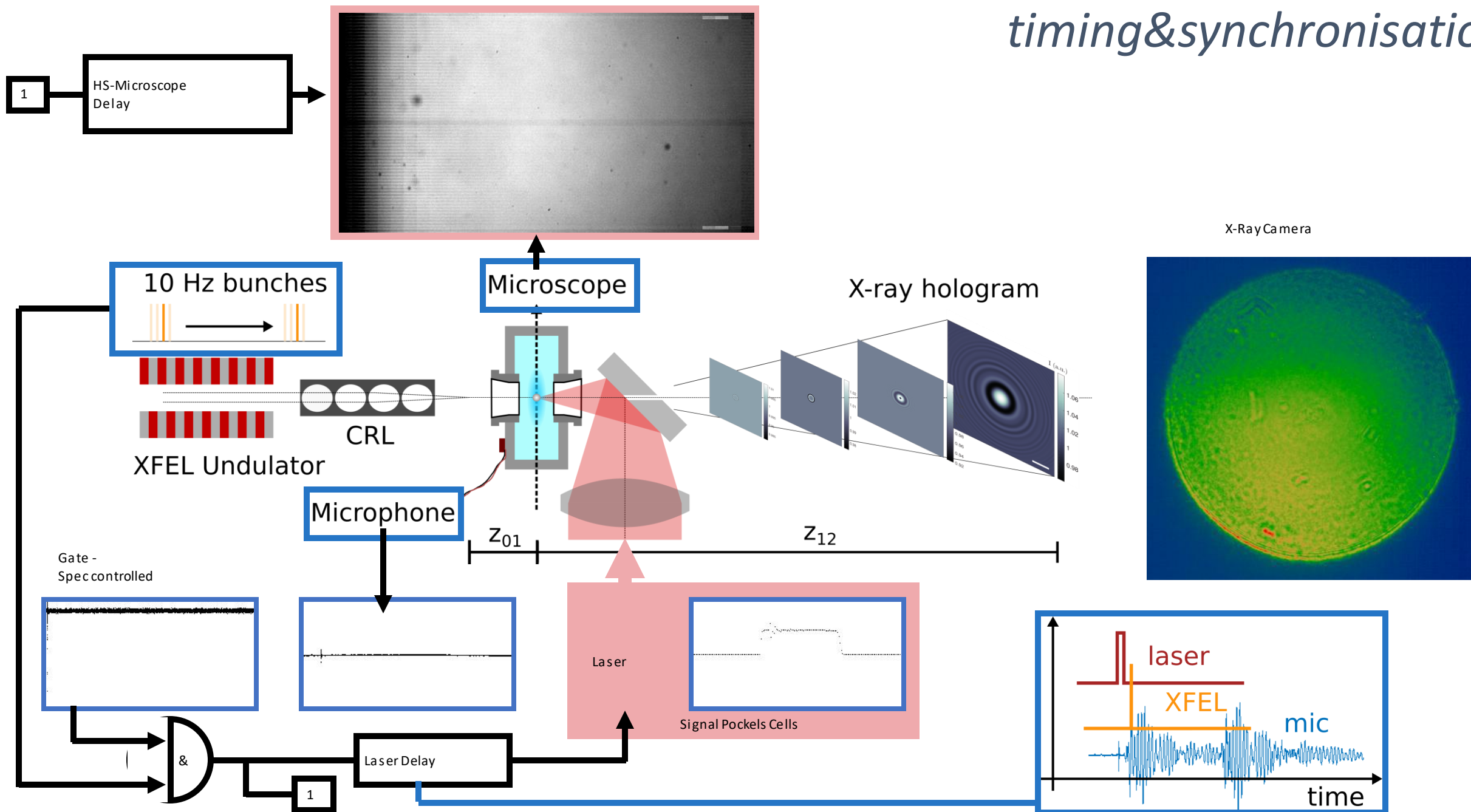


IR- alignment

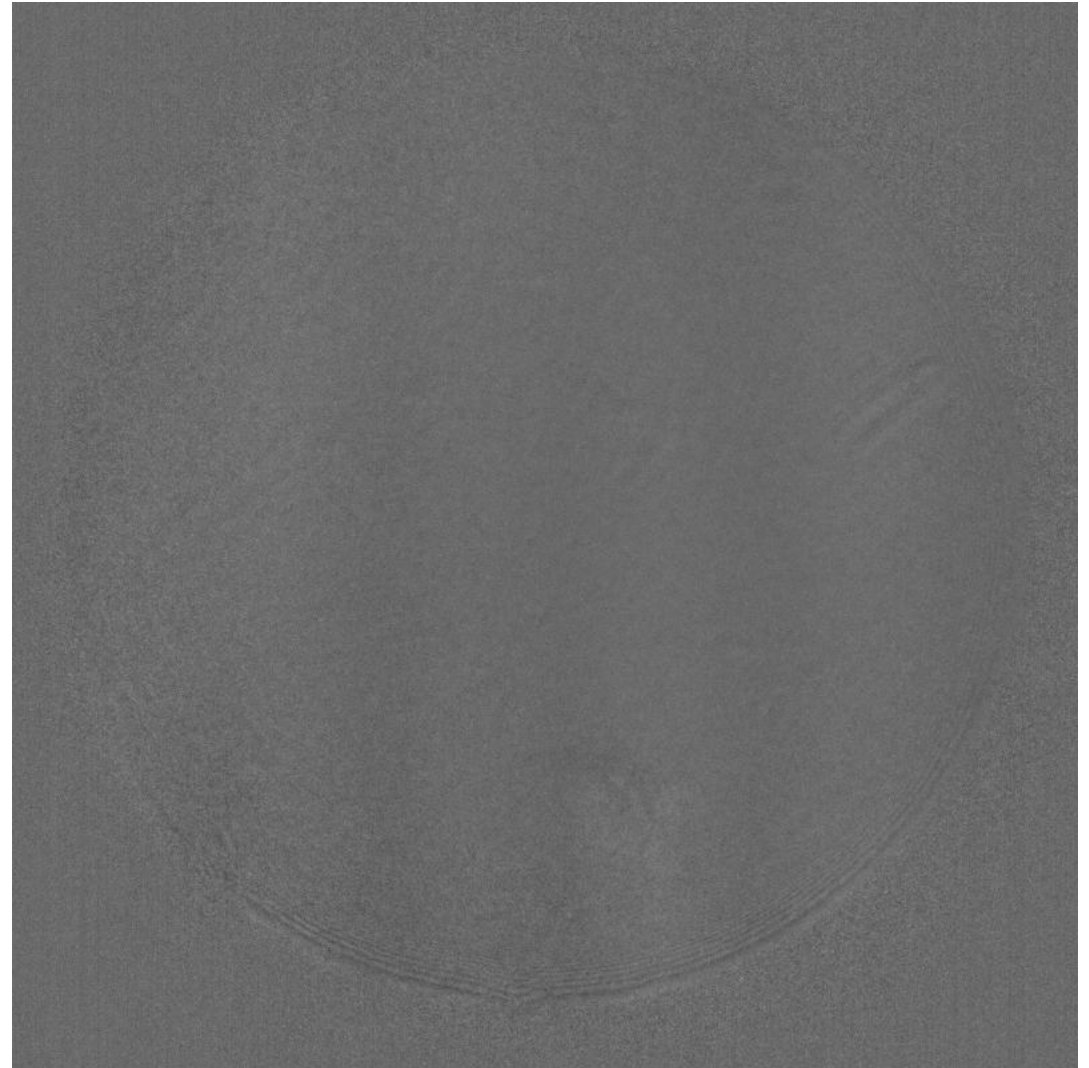
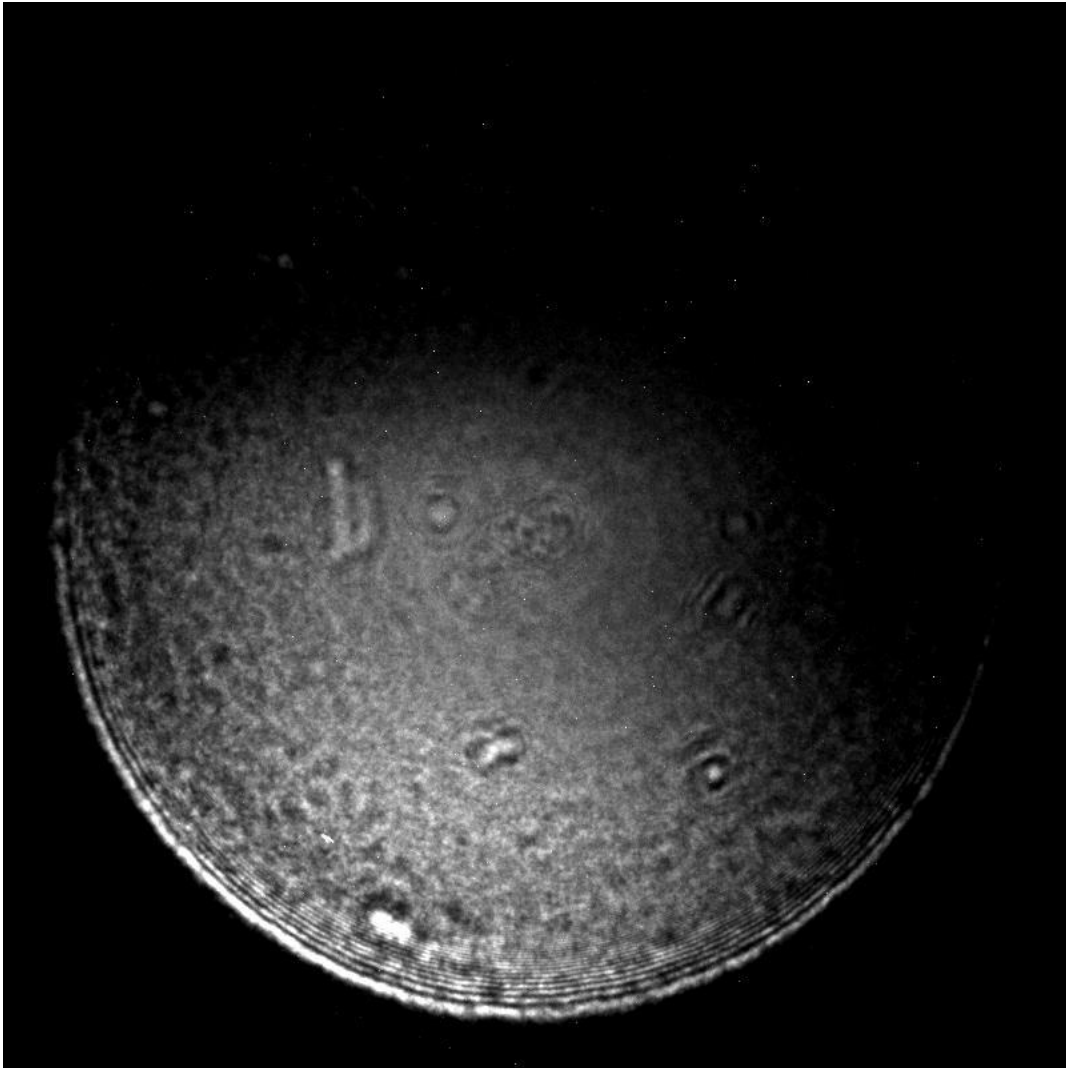
*you have
to have
spatial-
temporal
overlap !*

Malte Vassholz
Hannes Hoeppe

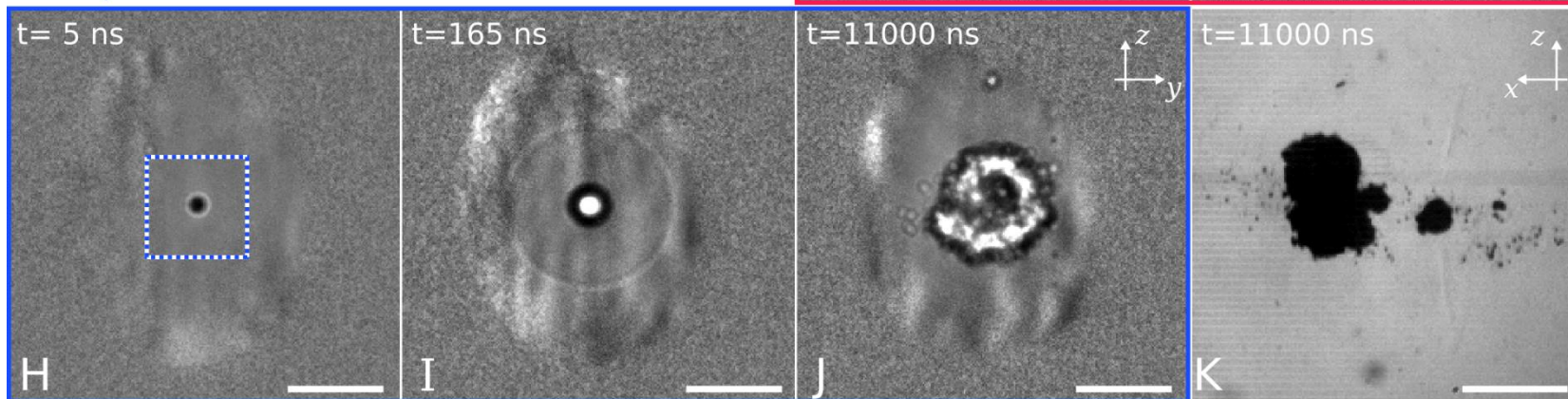
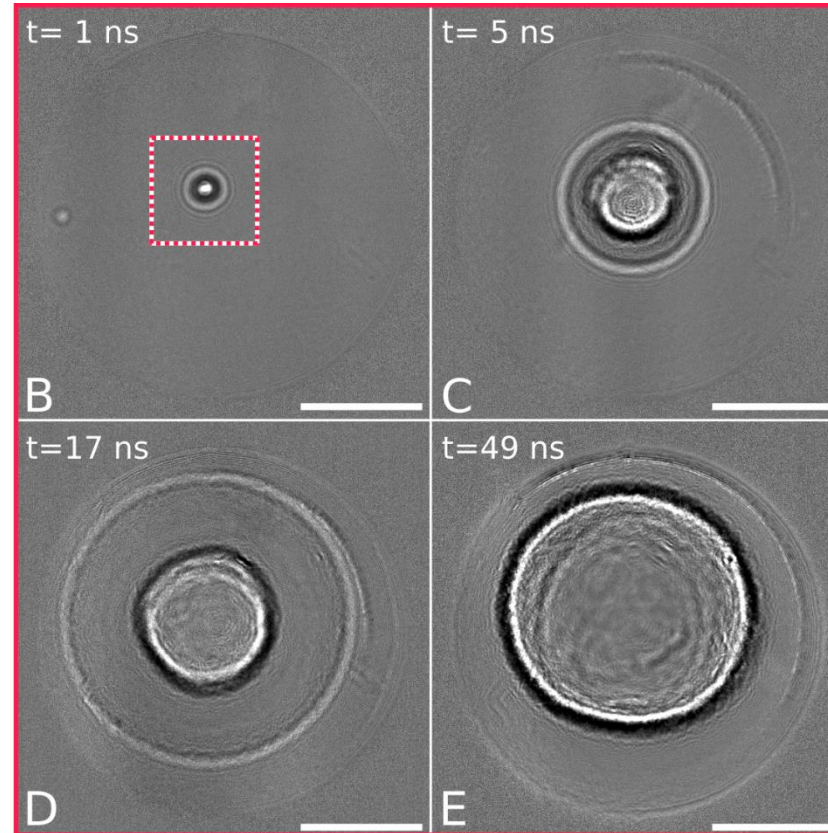
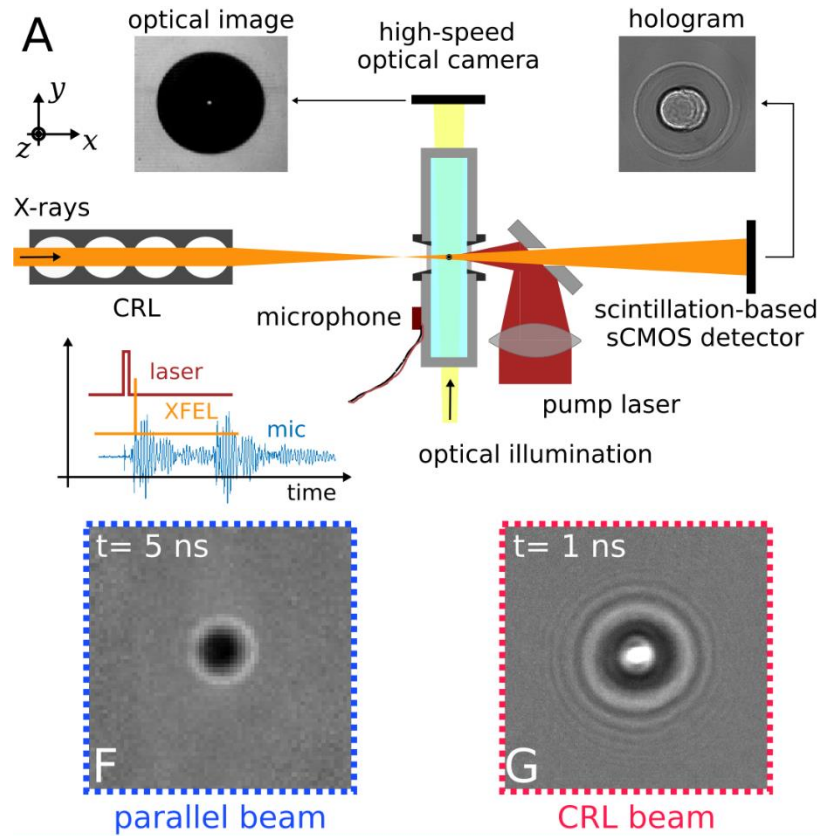
timing & synchronisation



This is what we saw when we saw something (online analysis)

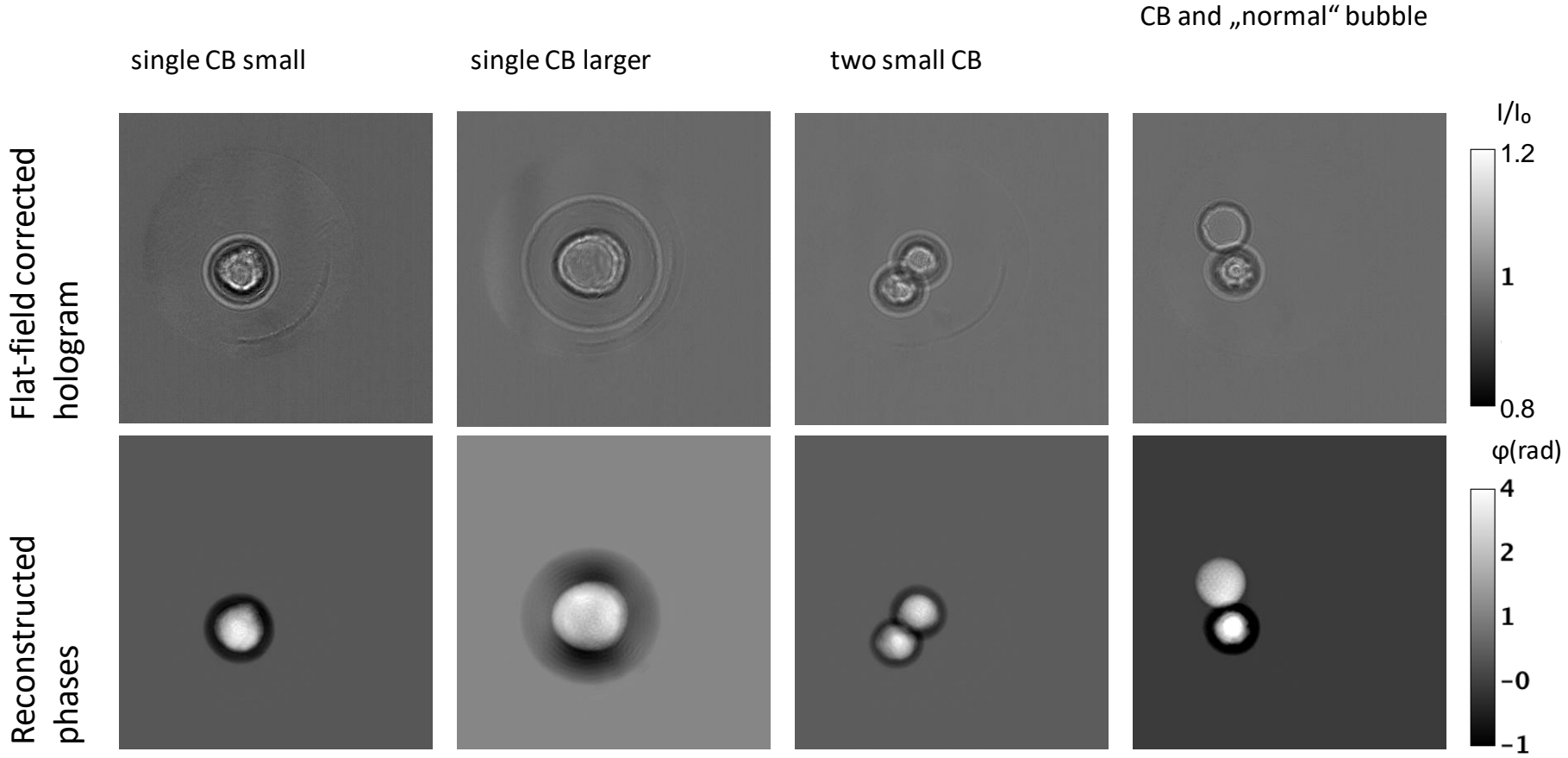


*o.k.
now we 're
talking !*

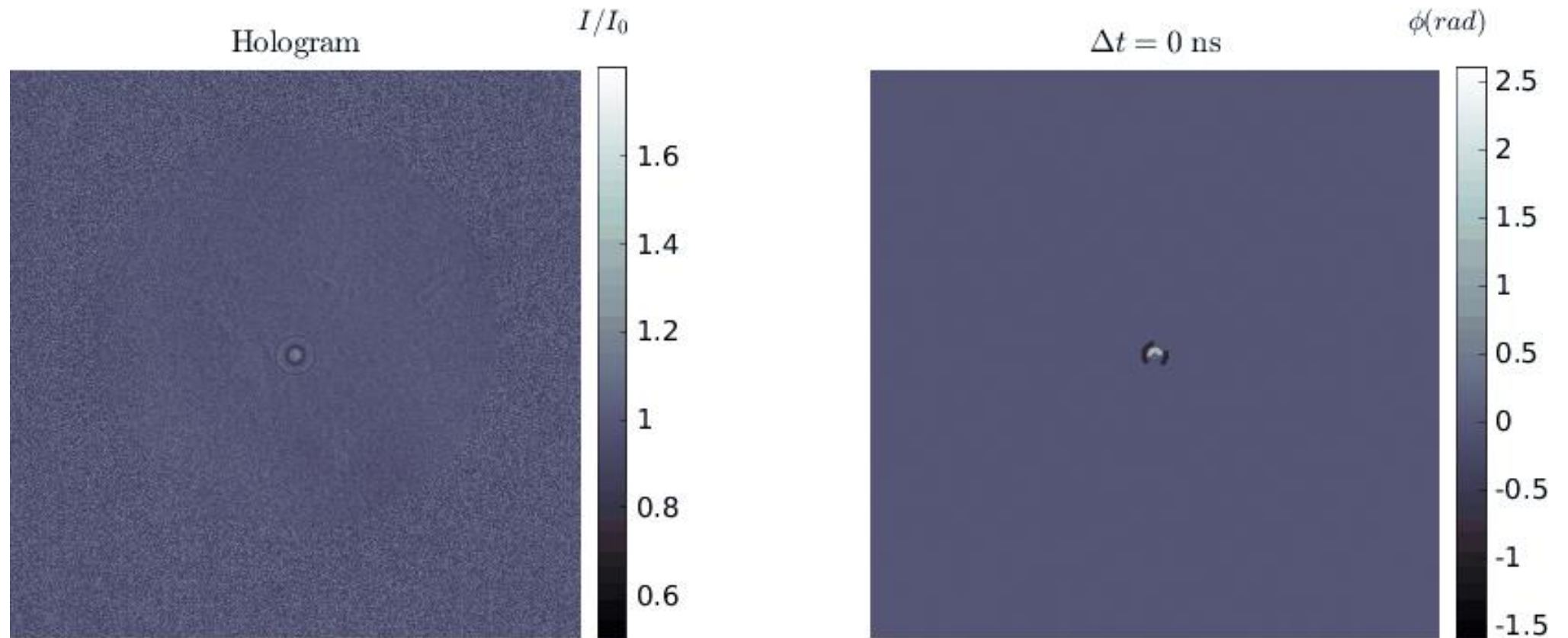


*But how to
analyze all
of this?*

Phase retrieval

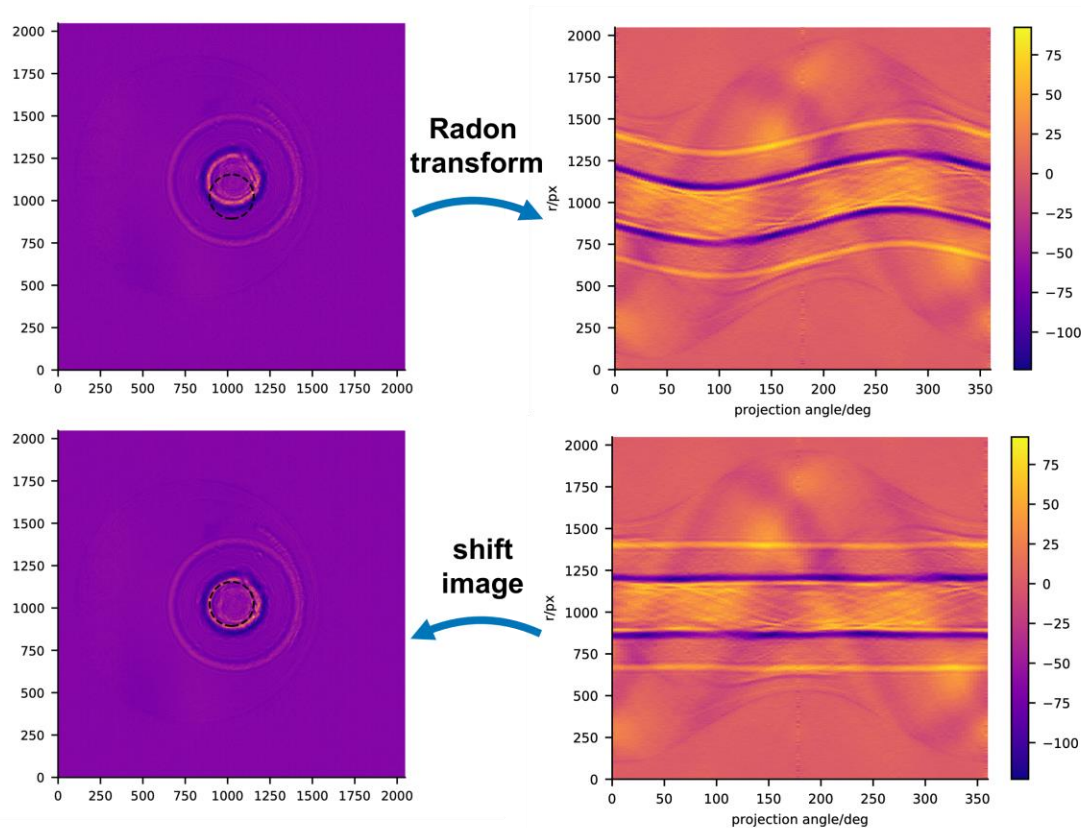


@ Fresnel number $Fr = 0.8 \cdot 10^{-3}$ (with respect to the pixel size)
pixel size $\Delta x = 109 \text{ nm}$, FOV = $140 \mu\text{m}$

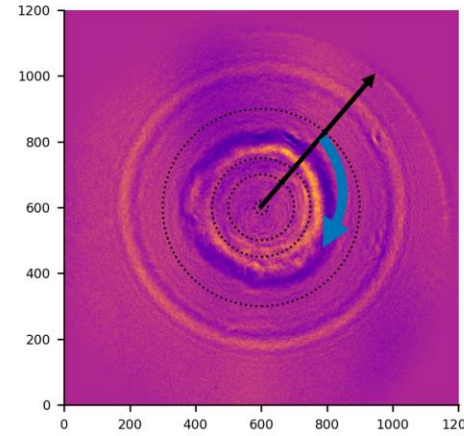


@ Fresnel number $Fr = 7.6 \cdot 10^{-4}$ (with respect to the pixel size) pixel size $\Delta x = 97.7$ nm, FOV = 140 μm

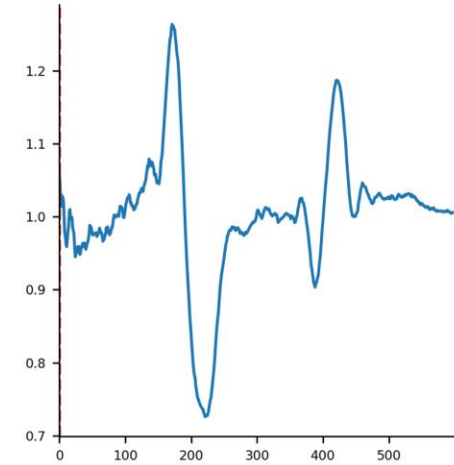
Propagation of radially symmetric wavefunctions & data analysis



Integrate over polar angle



radial intensity



$$\tilde{g}(k) = 2\pi \int_0^\infty g(r) J_0(rk) r dr$$

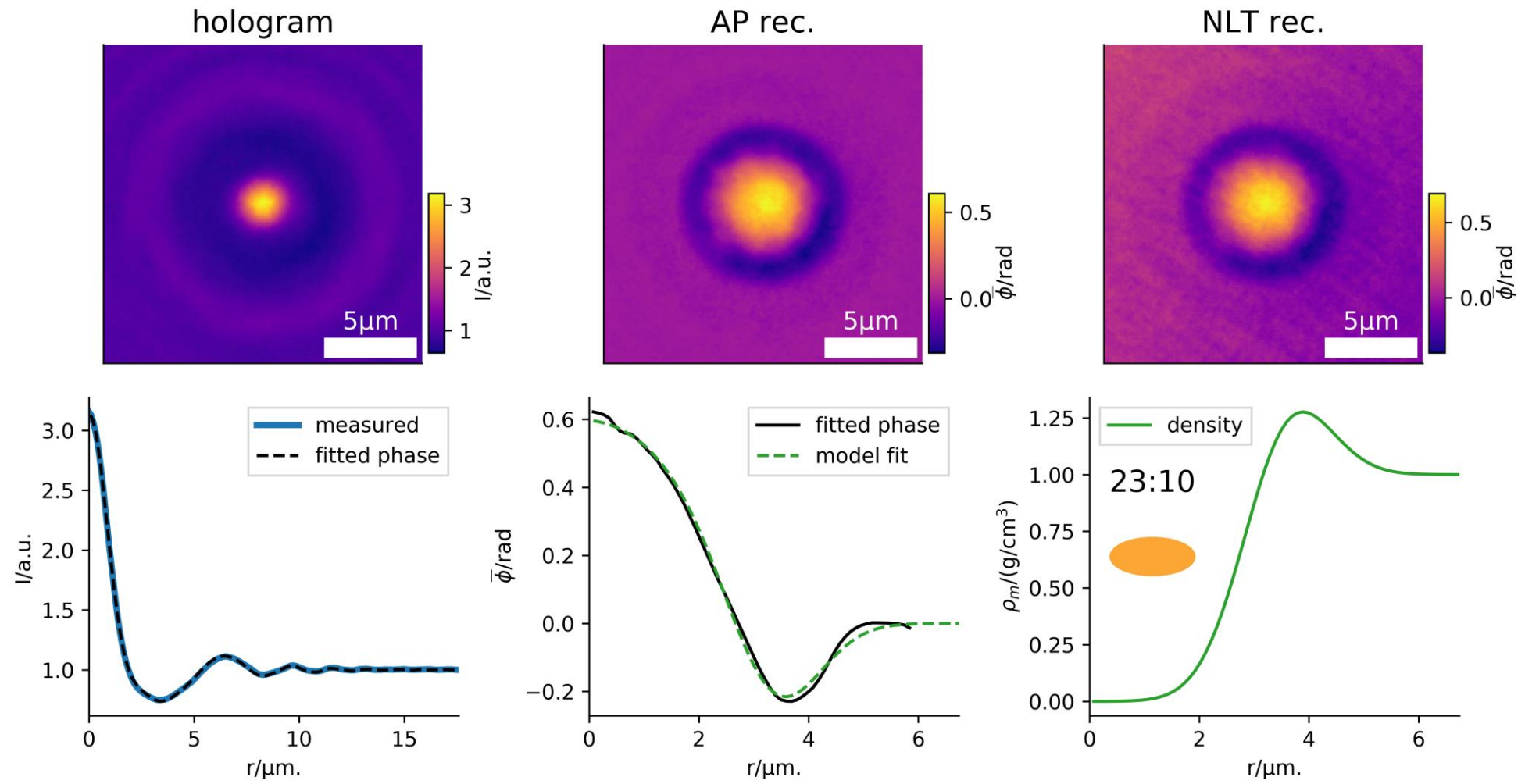
$$\tilde{g} = \mathcal{H}_0[g] \text{ Hankel transform with } \mathcal{H}_0^{-1} = \mathcal{H}_0$$

$$\psi(r_\perp, z) = \exp(ikz) \mathcal{H}_0 \left[\exp\left(\frac{-izk_\perp^2}{2k}\right) \mathcal{H}_0[\psi(r_\perp, 0)] \right]$$

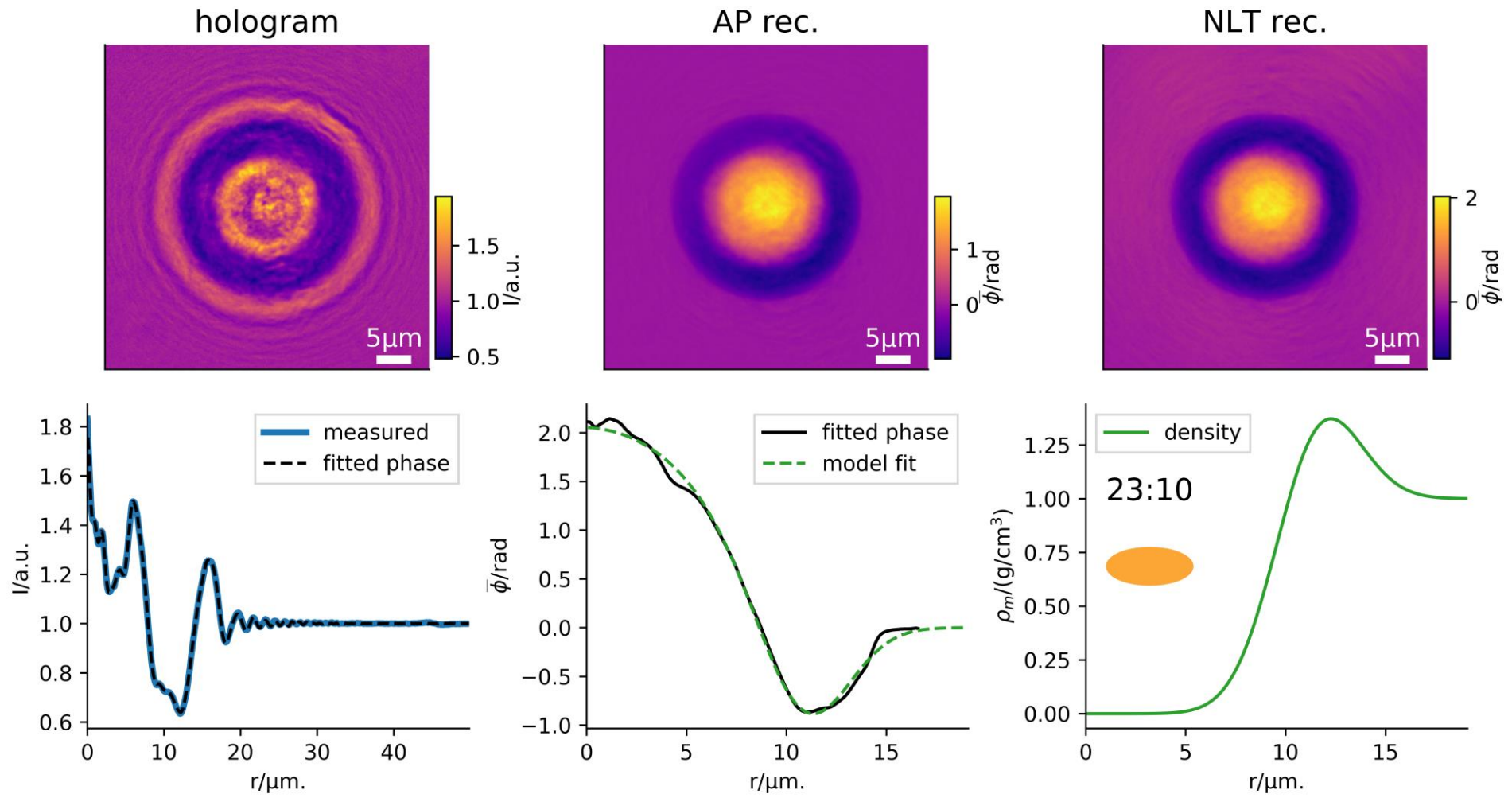
implemented on a discrete Hankel grid $\psi_i^{(z)} = M_{ij} \psi_j$

$\psi^* = \operatorname{argmin}(|I_{exp} - (M\psi)^2|^2)$ plus reg.term if needed

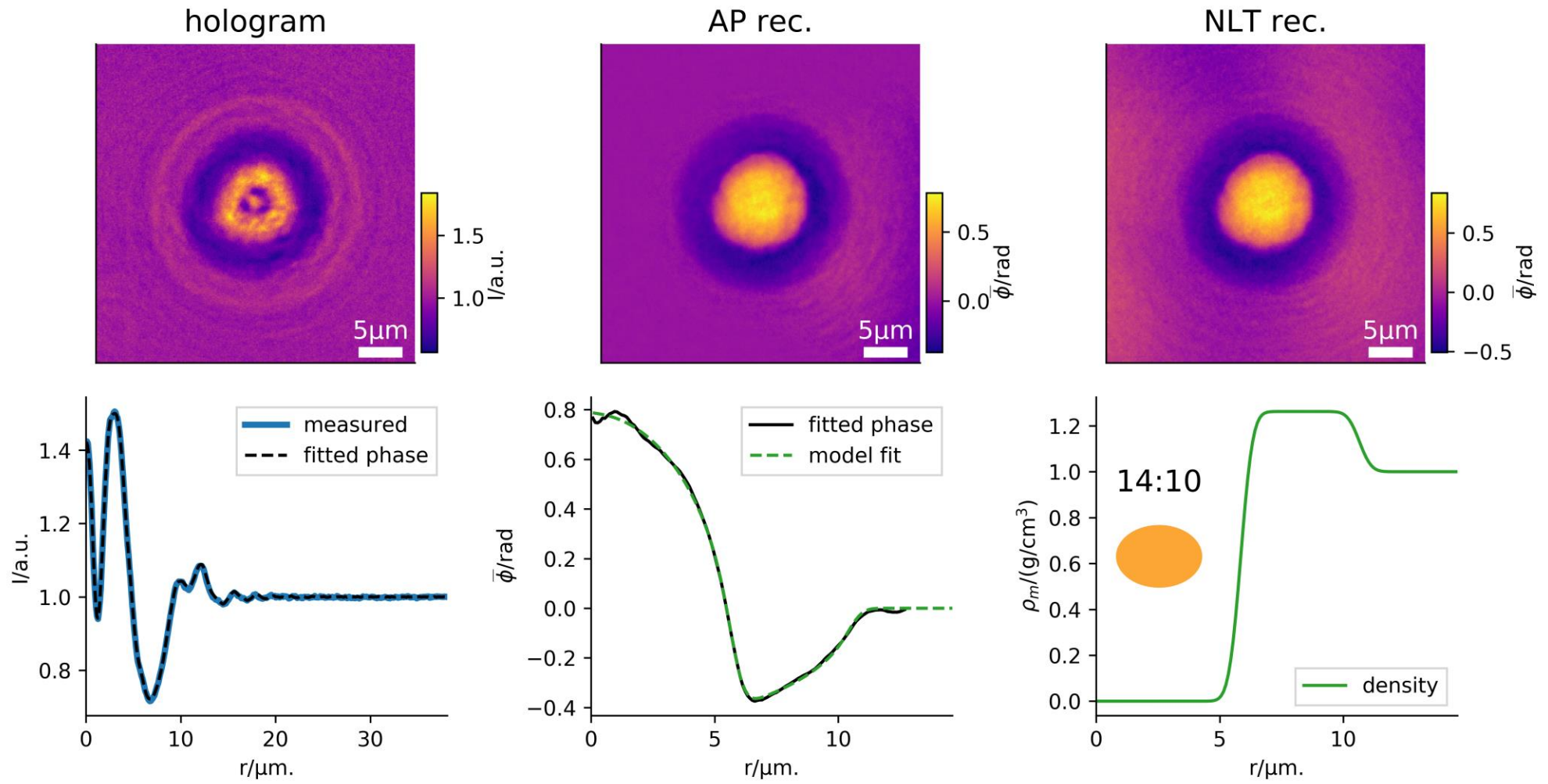
$\tau=1\text{ns}$



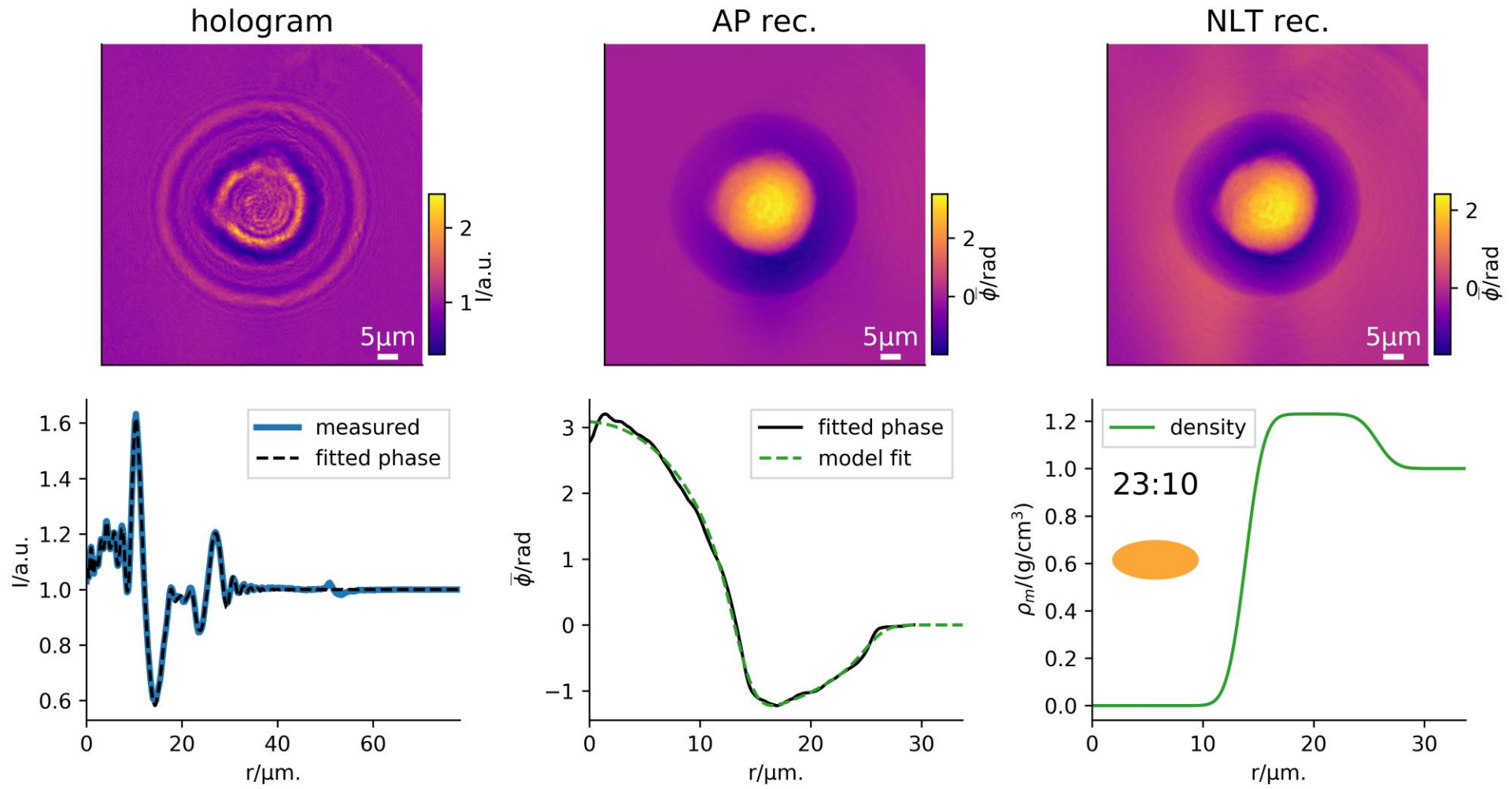
$\tau=4ns$



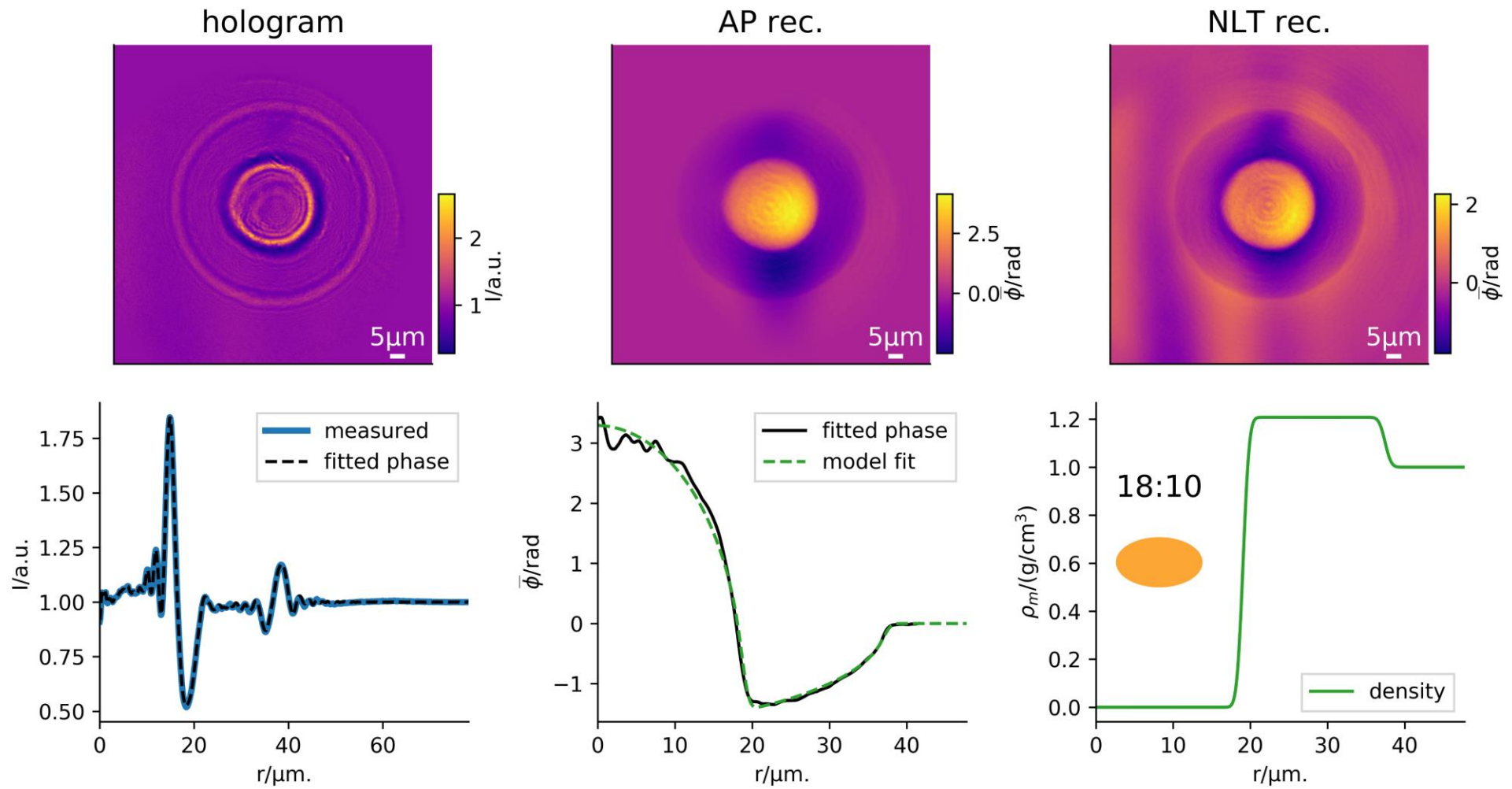
$\tau = 7\text{ns}$



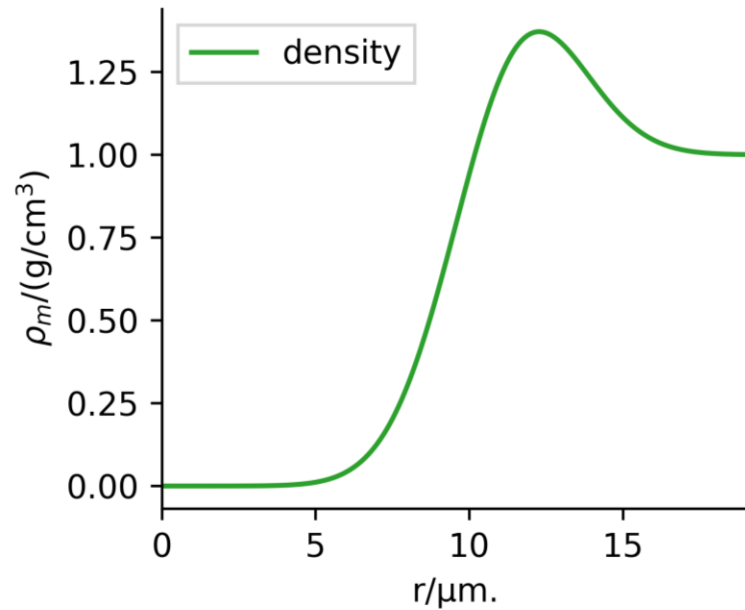
$\tau=10\text{ns}$



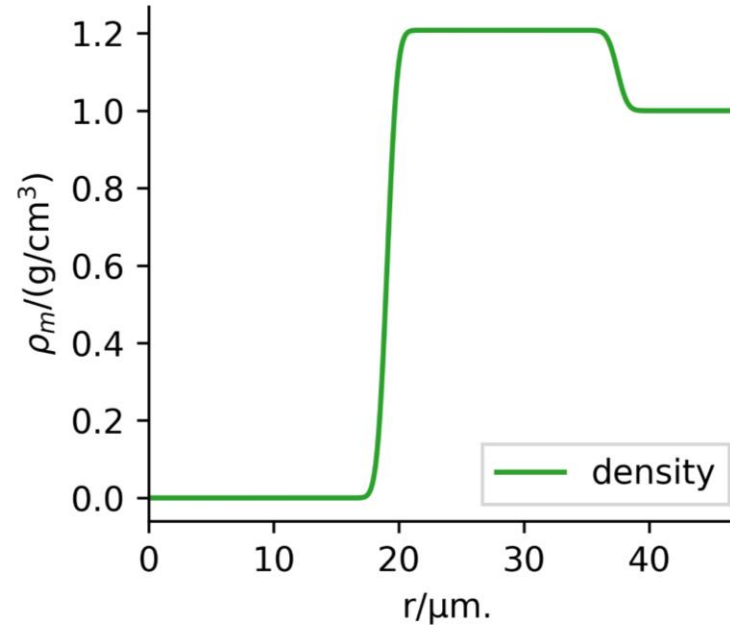
$\tau=13\text{ns}$



$\tau=4ns$



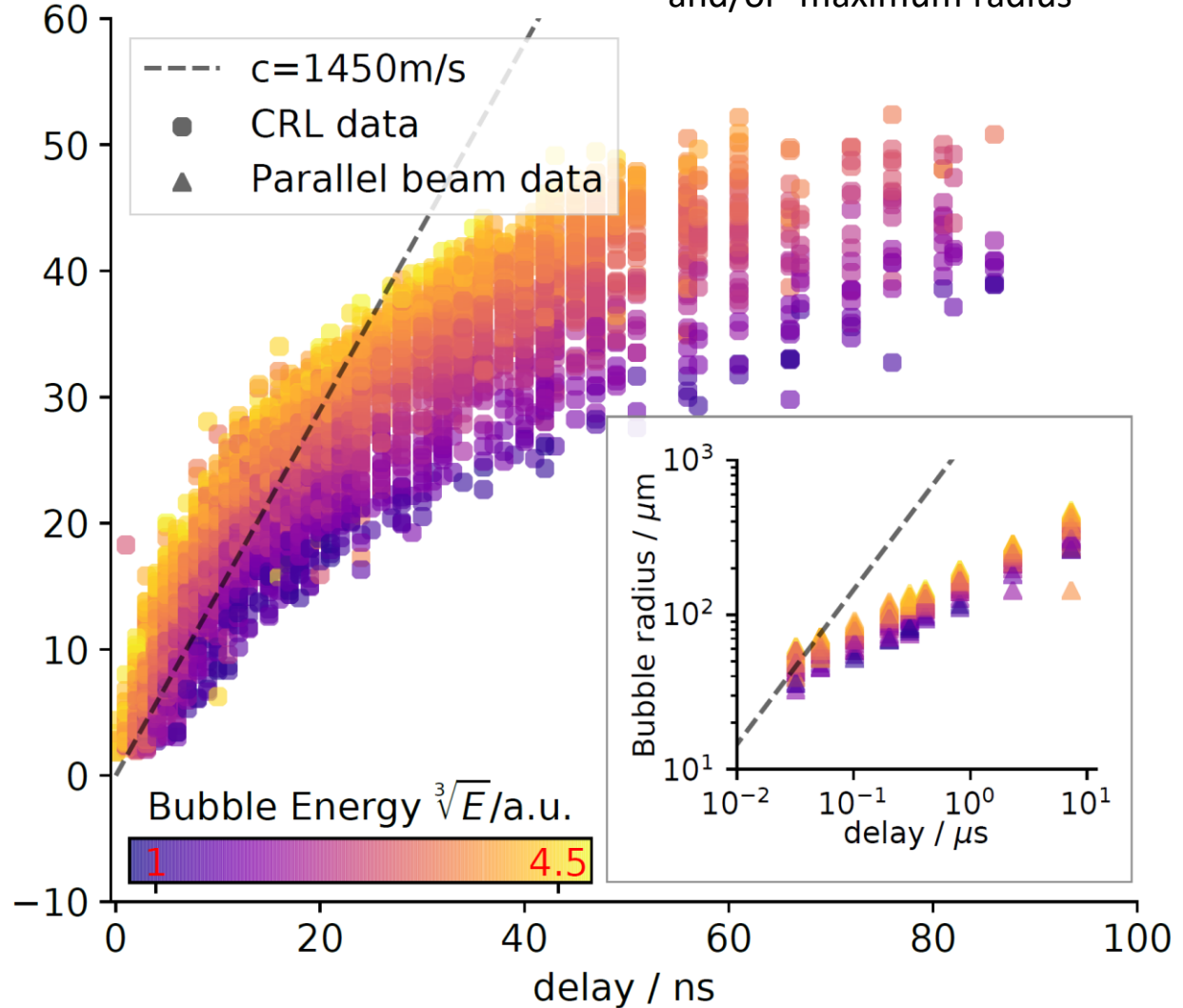
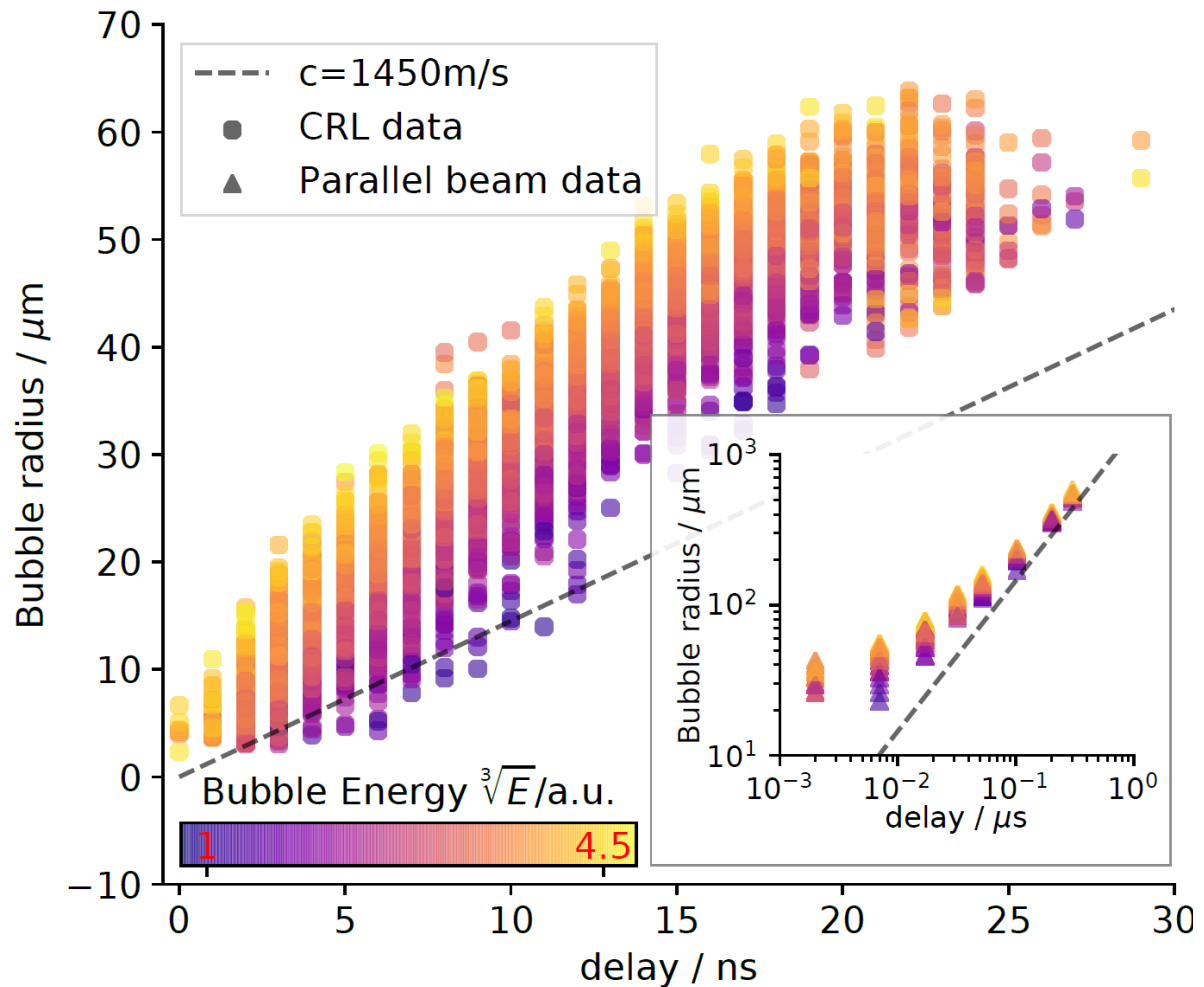
$\tau=13ns$



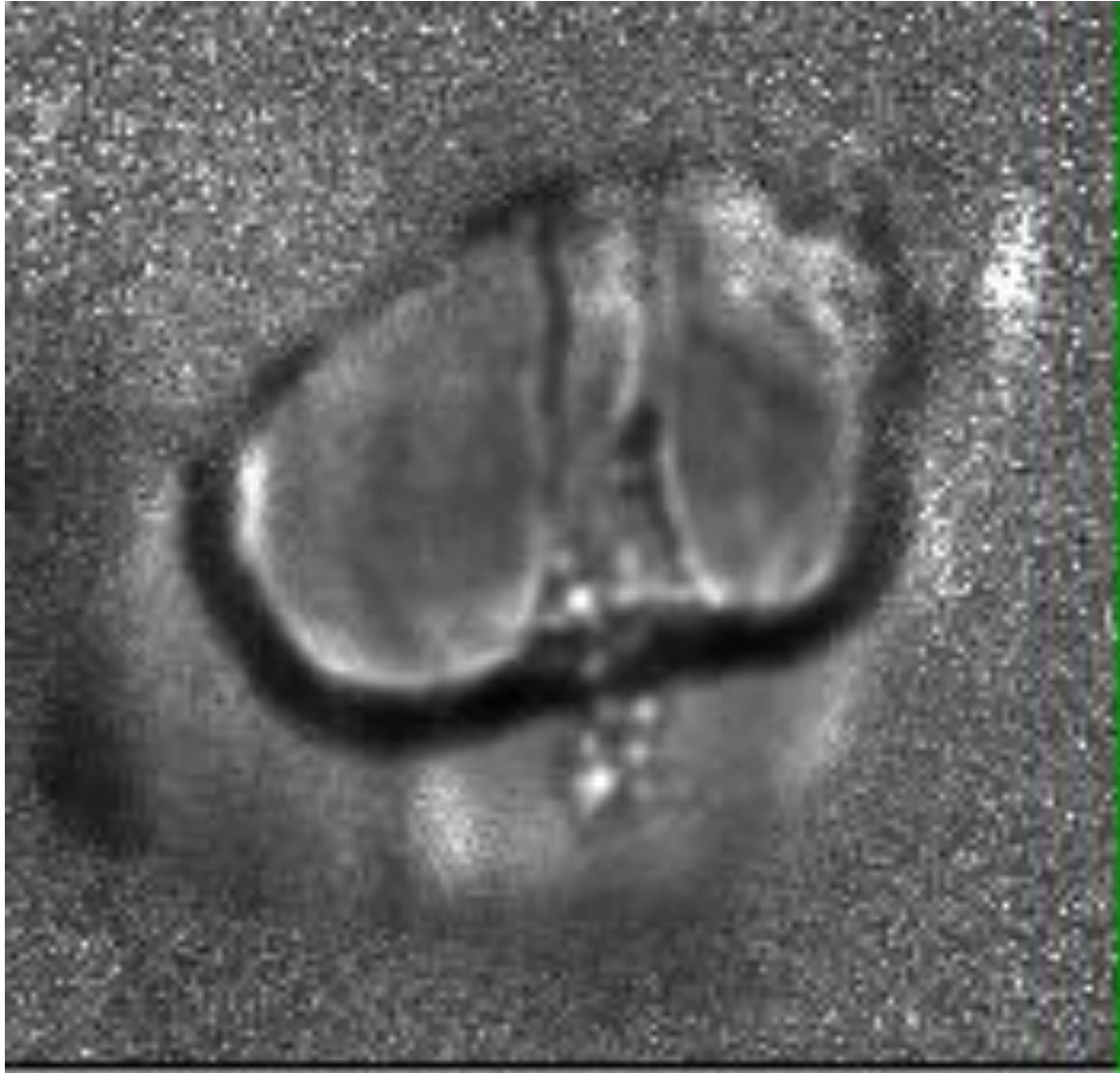
- *emergence of phase boundary / sharpening of the interface*
- *density of the shock wave -> equation of state*
- *Outlook: comparison with MD simulation*
- *Outlook: fs time scales / plasma dynamics / water structure in shock wave collapse of the bubble*

Working out the entire ensemble / sorting for E_B

- maximum radius (photon)
- life time (microphone)
- bubble energy from life time and/or maximum radius



This is not only about spherical bubbles...jetting

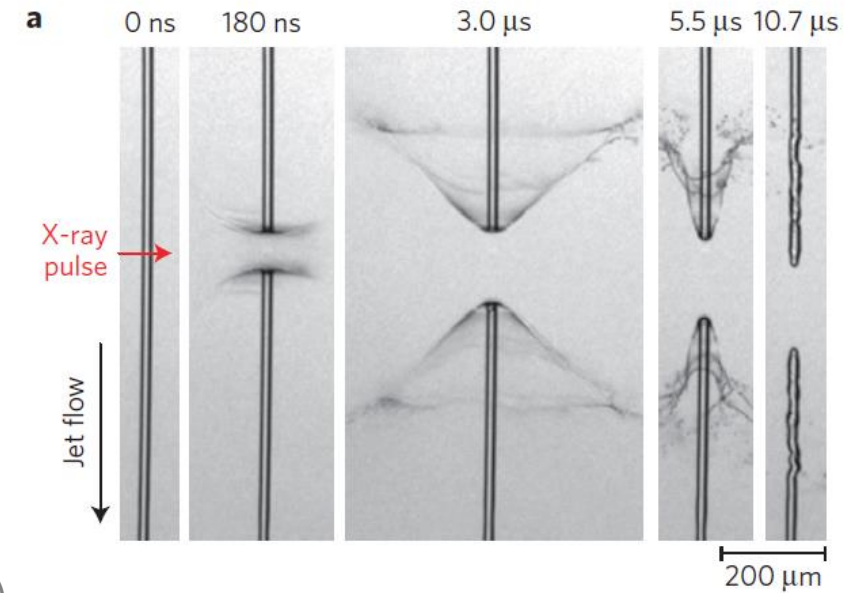
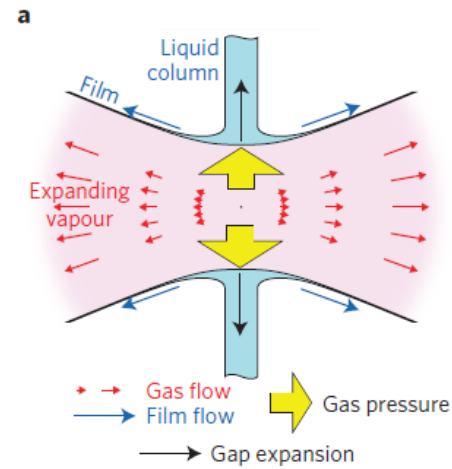


- cavitation bubbles near surfaces
- jetting
- parallel beam illumination

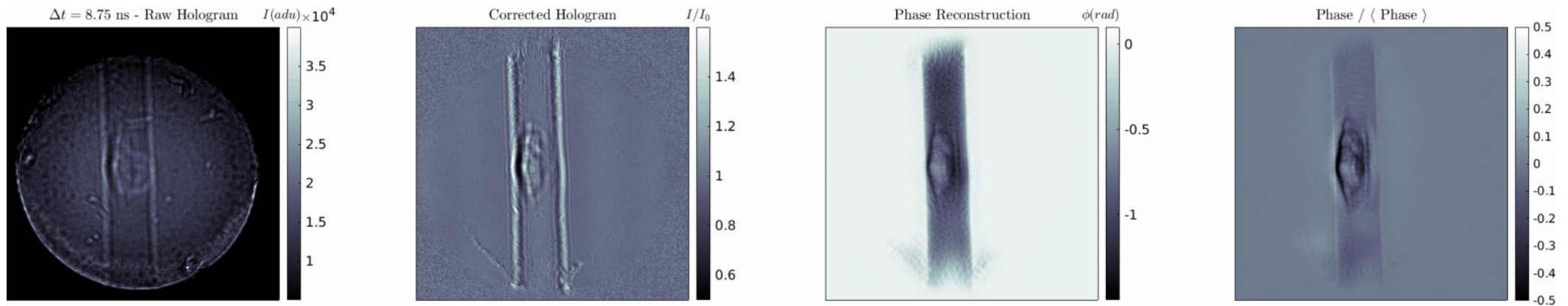
Cavitation in the microfluidic jet

we can now observe the inverse

- IR pump / x-ray probe
- density profiles
- Outlook: extend fs-regime



Stan et al. Nat.Phys (2016)



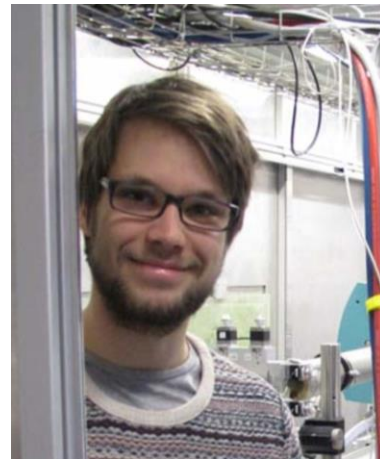
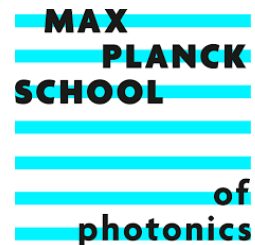
Small bubble collaboration & Acknowledgements

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Universität Göttingen, III-Phys. Inst.-Biophysik & Institut für Röntgenphysik

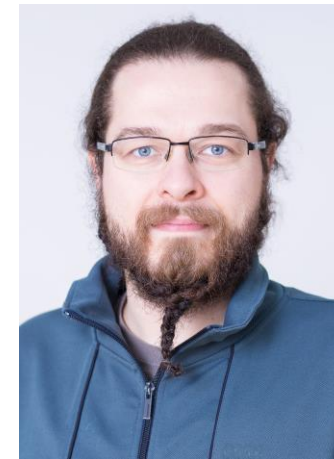
Johannes Hagemann, Frank Seiboth, Andreas Schropp, Christian G. Schroer,
DESY Photon Science

Johannes Möller, Jörg Hallmann, Ulrike Boesenberg, Chan Kim, Markus Scholz, Alexey Zozulya, Wei Lu,
Roman Shayduk, Robert Schaffer, Anders Madsen
MID / XFEL

- UAC MID 06/2019 @14keV
- Beamtime 10/ 2019 @17.8keV
(p2207 & p2545 Hagemann/Salditt)



Vassholz *et al.*



Hagemann *et al.*

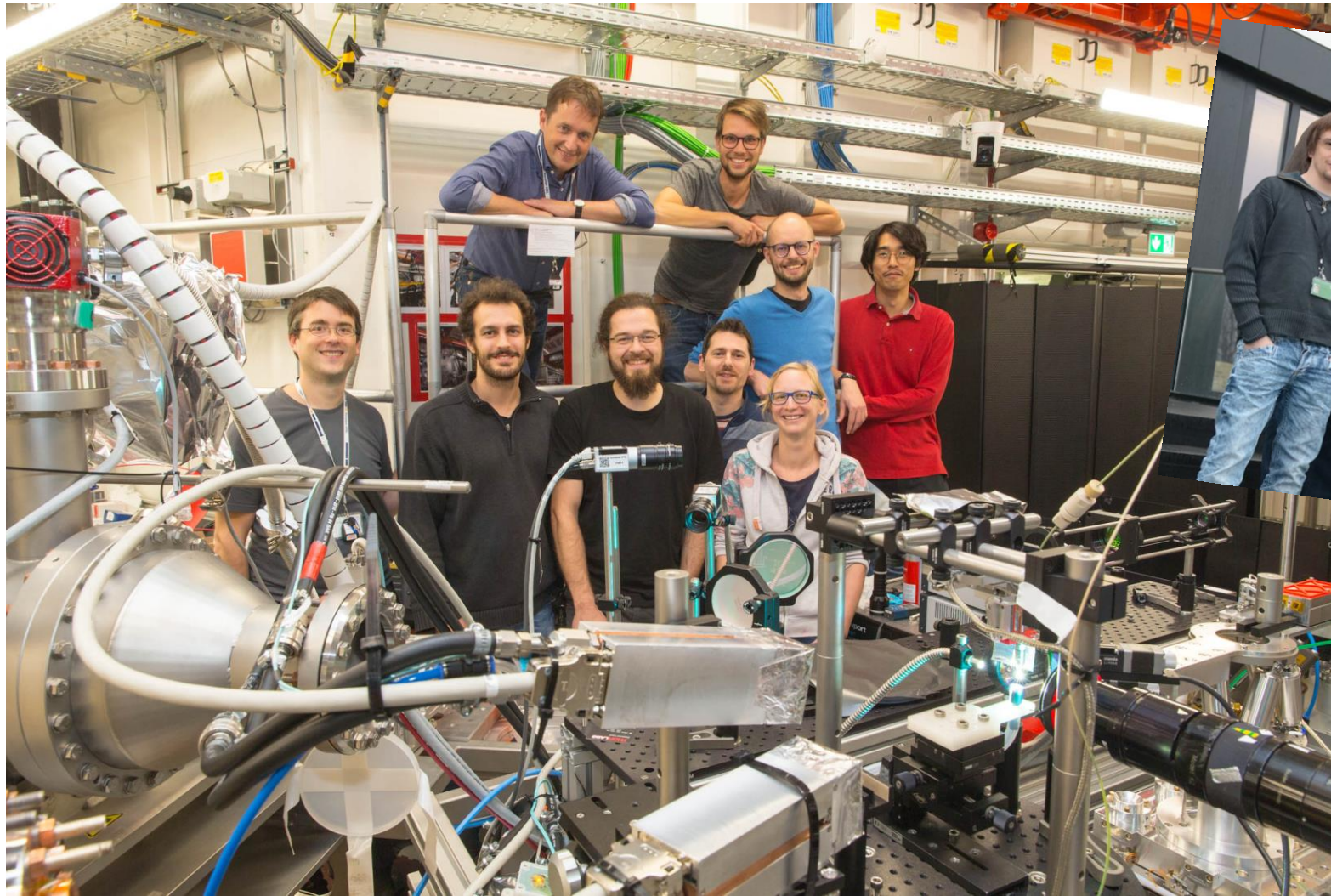


Osterhoff *et al.*

& the review panel for their trust!

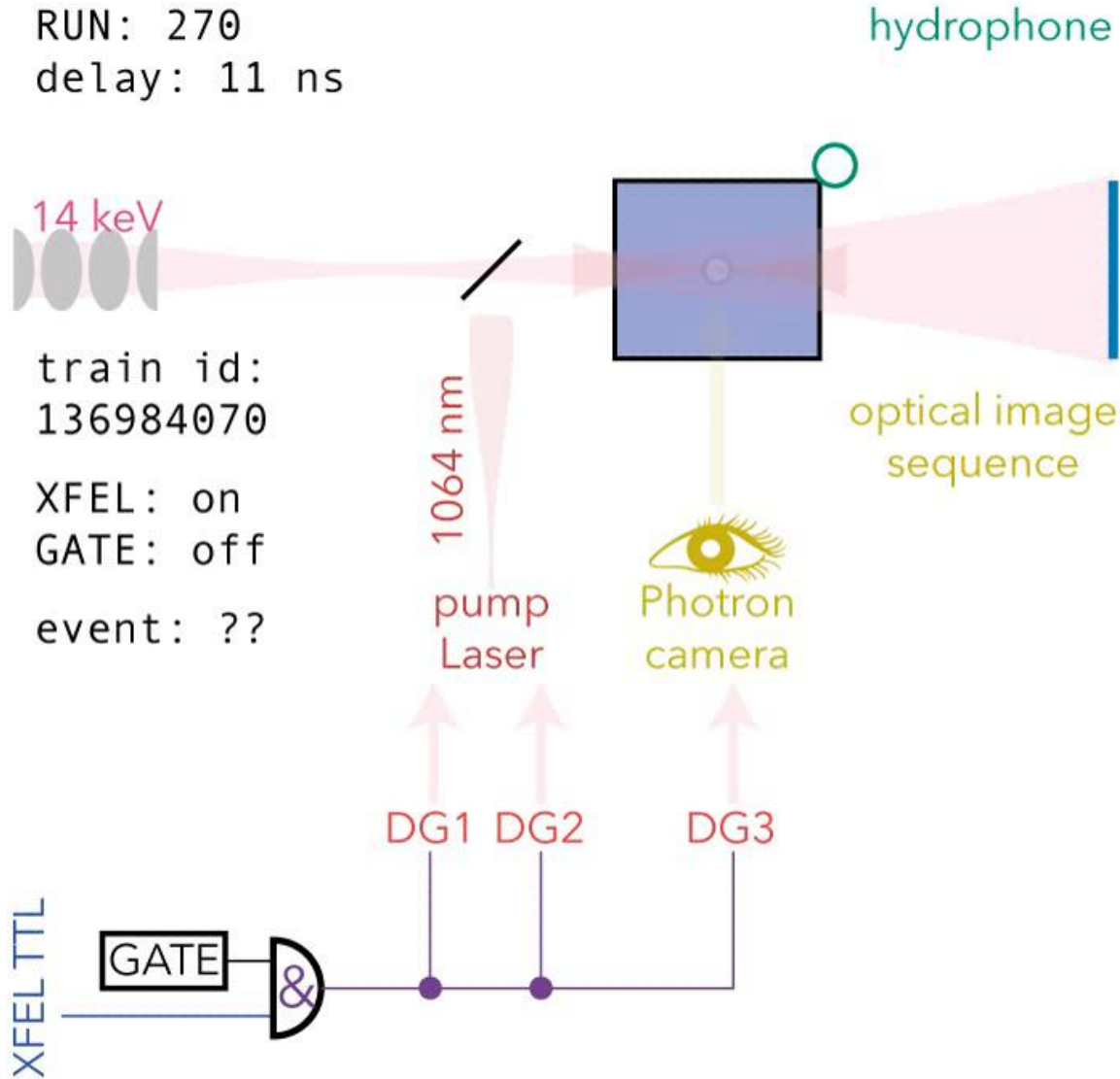
Manuscripts in preparation

Small bubble collaboration...



Many thanks to the MID team
for an amazing instrument
And fruitful collaboration

timing&synchronisation



X-ray raw data

hologram

phase image

Talk to the experts @ the posters!

#30 Timing (M. Osterhoff)
#249 Imaging (M. Vassholz)
#255 Dynamics (H. Hoeppe)