Time-resolved X-Ray Holography

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Holography

Dennis Gábor 1900 – 1979

(1921-24 @ TUB)

Nobel Prize Physics

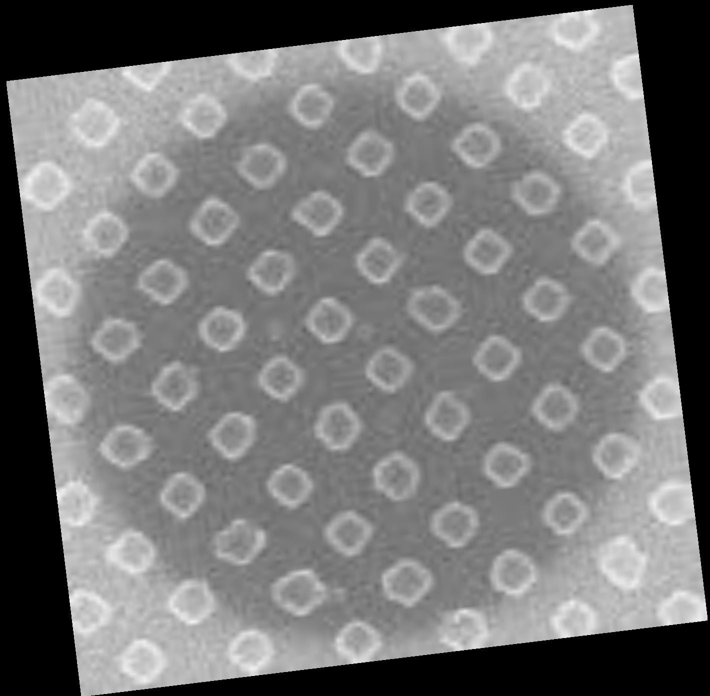
Holography and 3D information

Picture of analog hologram reconstruction from two angles

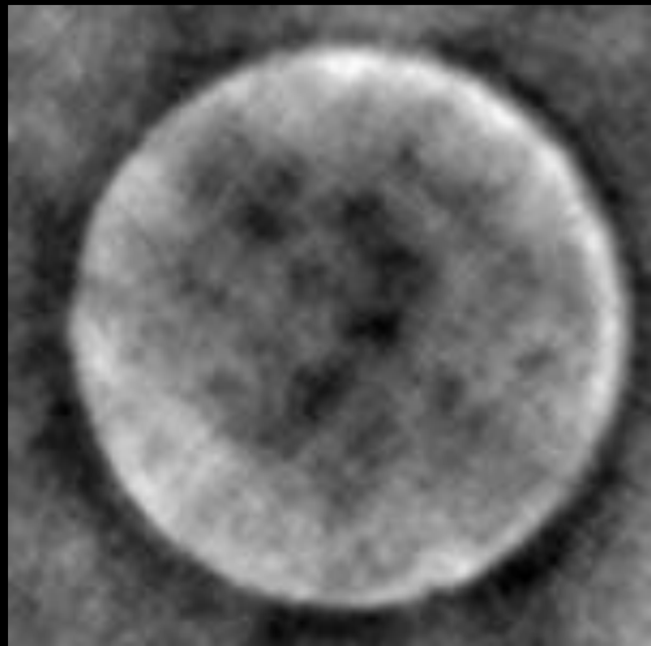
Teaser: see magnetization on the nanoscale via holography

Mesa Structures, 80 nm, 200 nm pitch, coated with a ferromagnetic thin film

Topography (SEM)

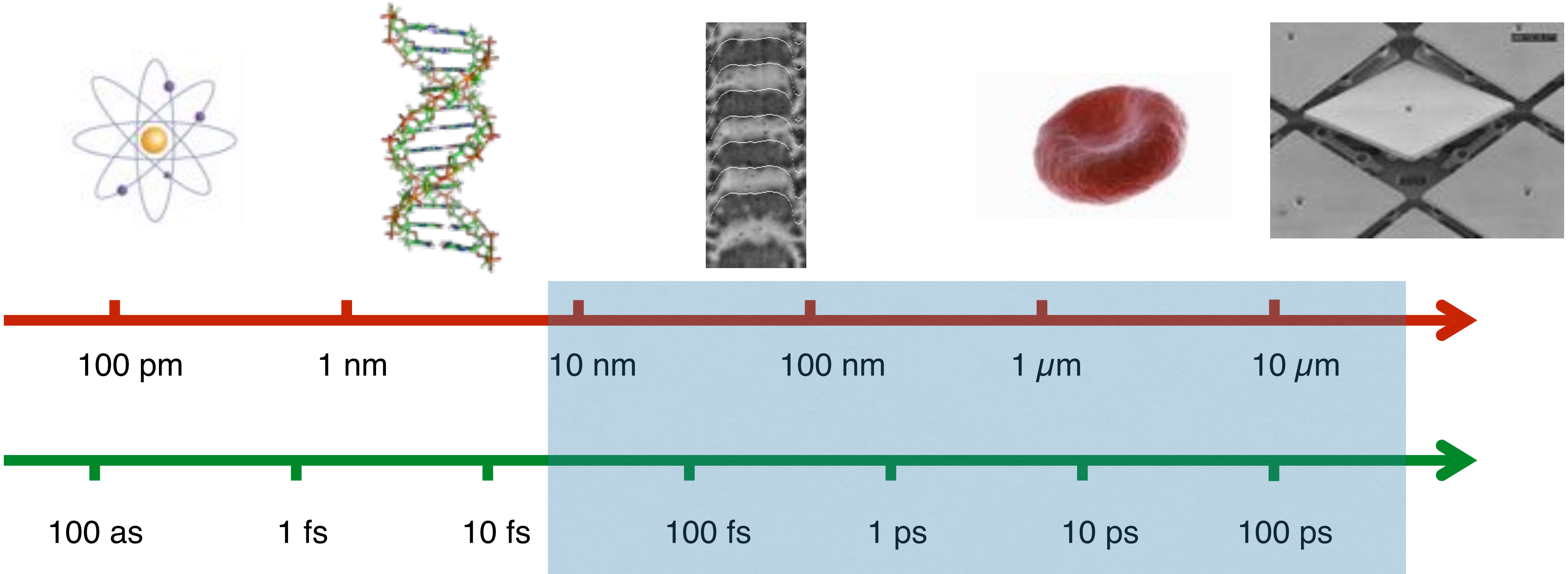


Magnetization



- How does one obtain (magnetic) contrast ?
- How can an image be generated ?
- How can dynamic processes be captured ?

Length and time scales in condensed matter



Electron dynamics within atoms

Ballistic electron movement

Atomic / molecular vibrations

Free electron motion in solids

Precession of a spin

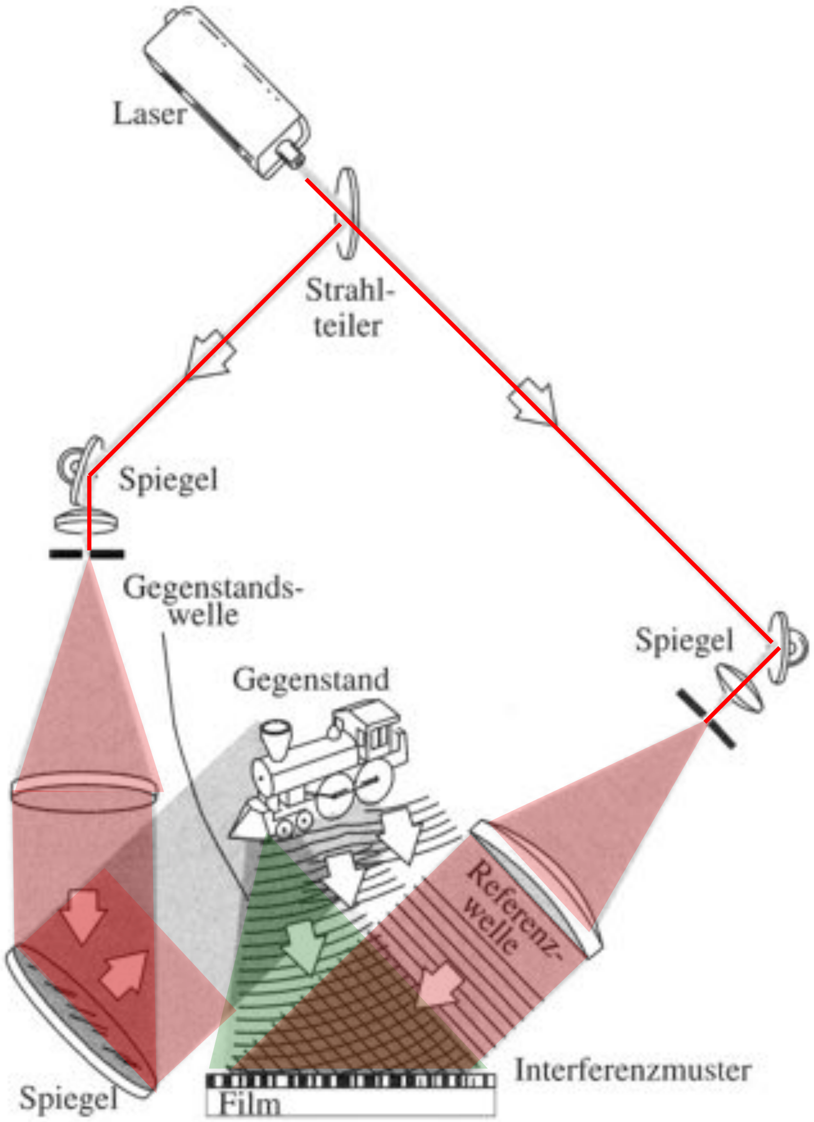
HDD magnetic recording time

Study dynamics (space & time) to understand principal mechanisms and macroscopic functionality
→
need to be sensitive to the entities you want to study (**contrast**) at the required **spatial** and **temporal resolution**

issues with (soft) x-rays

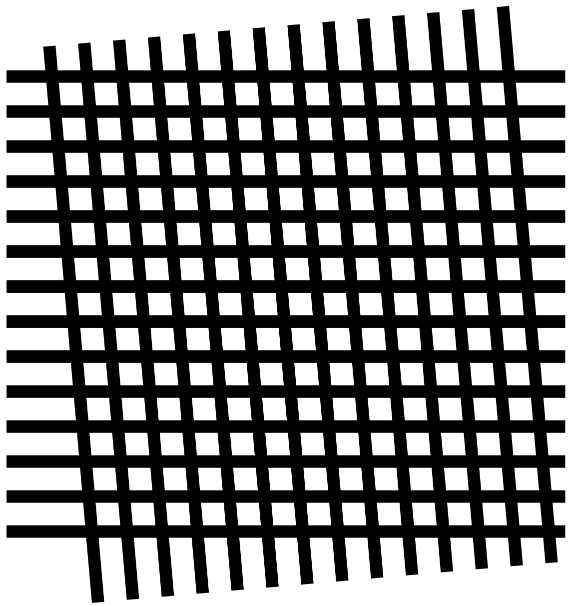
Max-Born-Institut

Holography with visible light



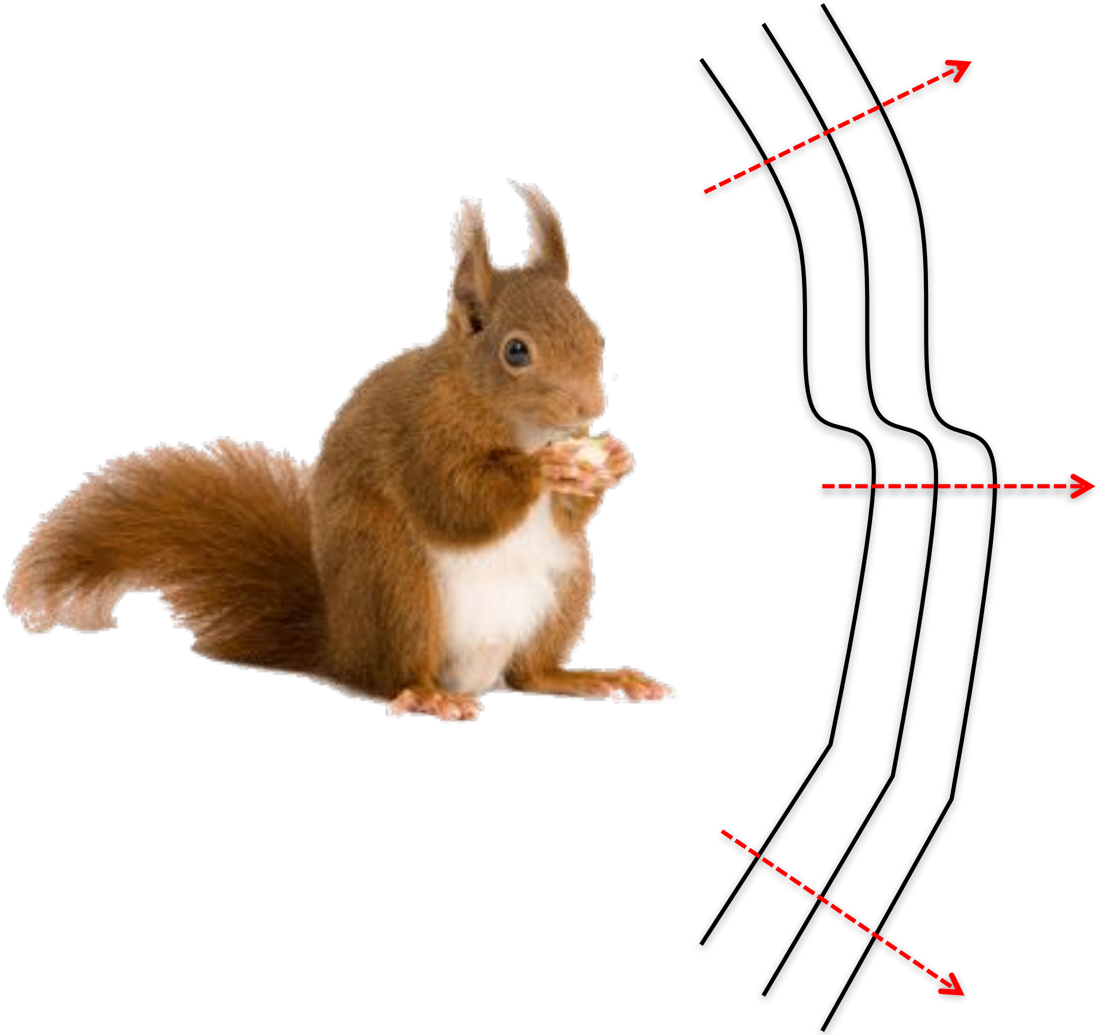
Problems with (soft) x-rays

- Efficient beam splitting is difficult
- In particular, reflectivities are only significant for very small glancing angles
- mechanical stability on the order of λ required
- Interference fringes have a separation of ≈λ unless included angle becomes small



Encoding the exit wave

in the holographic measurement, we can encode the **wavefield (i.e. amplitude and phase)** of the light leaving the object over a certain solid angle



- If that is known, we can **calculate** how the wavefield propagates forward (in vacuum or a homogeneous medium).
- Alternatively, this **just happens** when we carry out an analog reconstruction of the image via suitable illumination

Why x-ray holography?

X-ray holography is a high-resolution imaging technique with some unique characteristics.

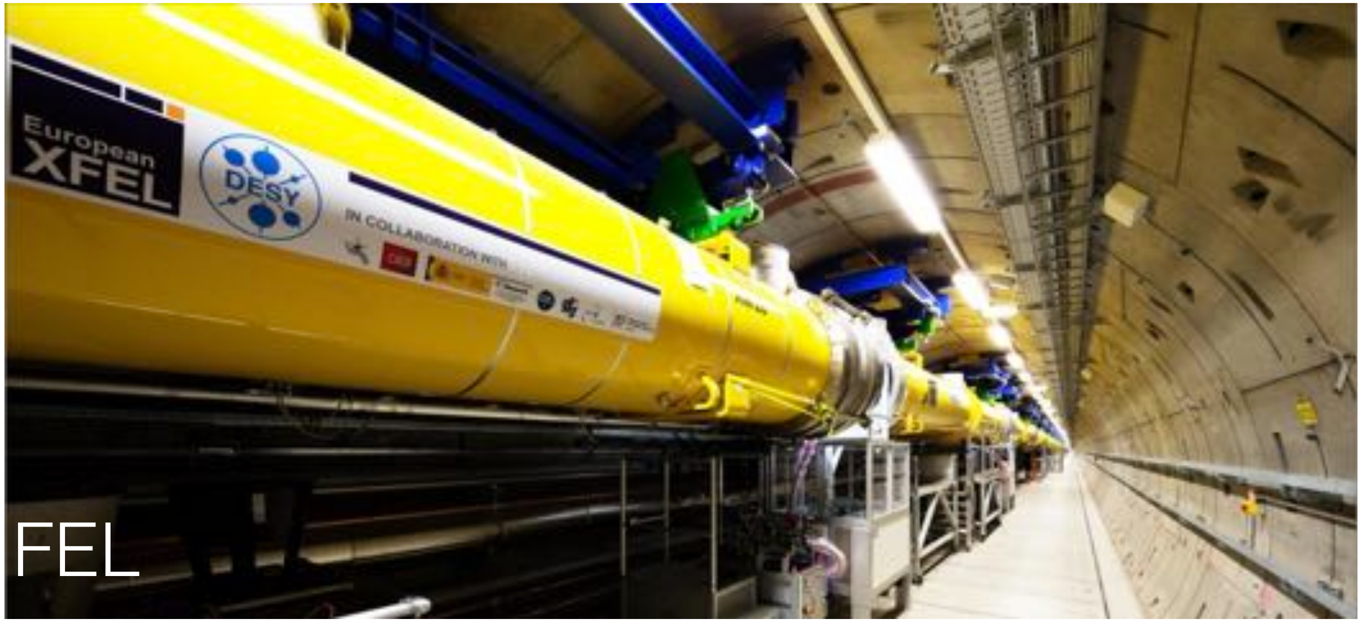
Why now?
Because we can:

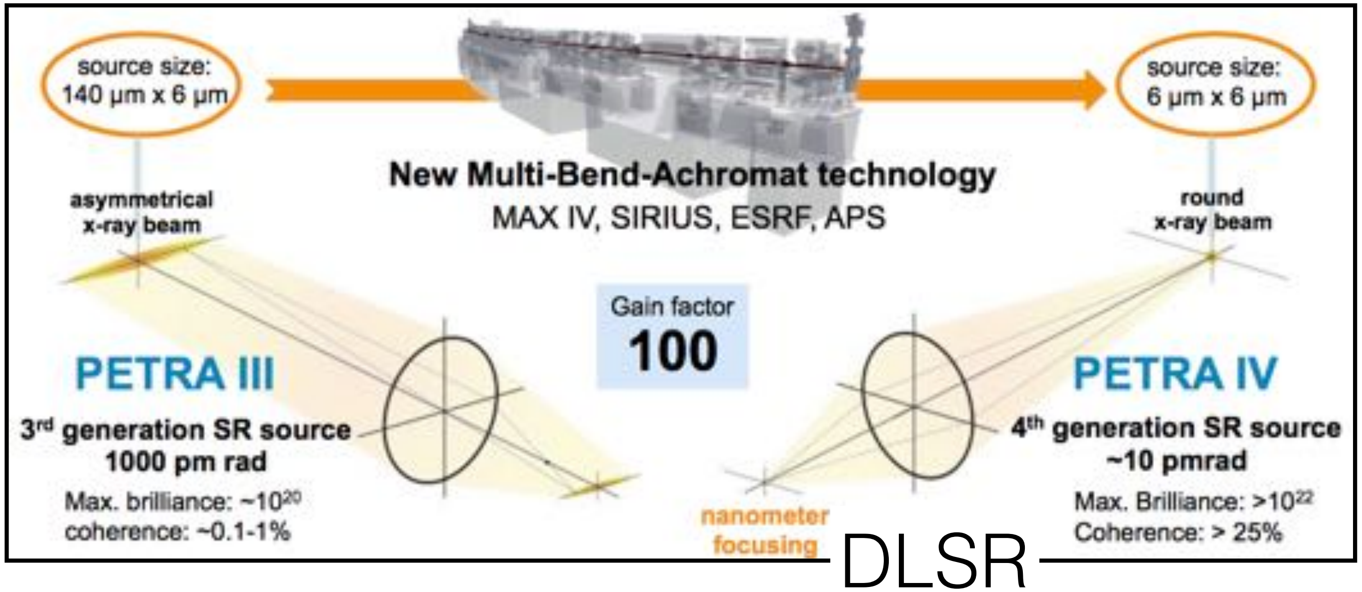
coherent x-ray sources are increasingly available

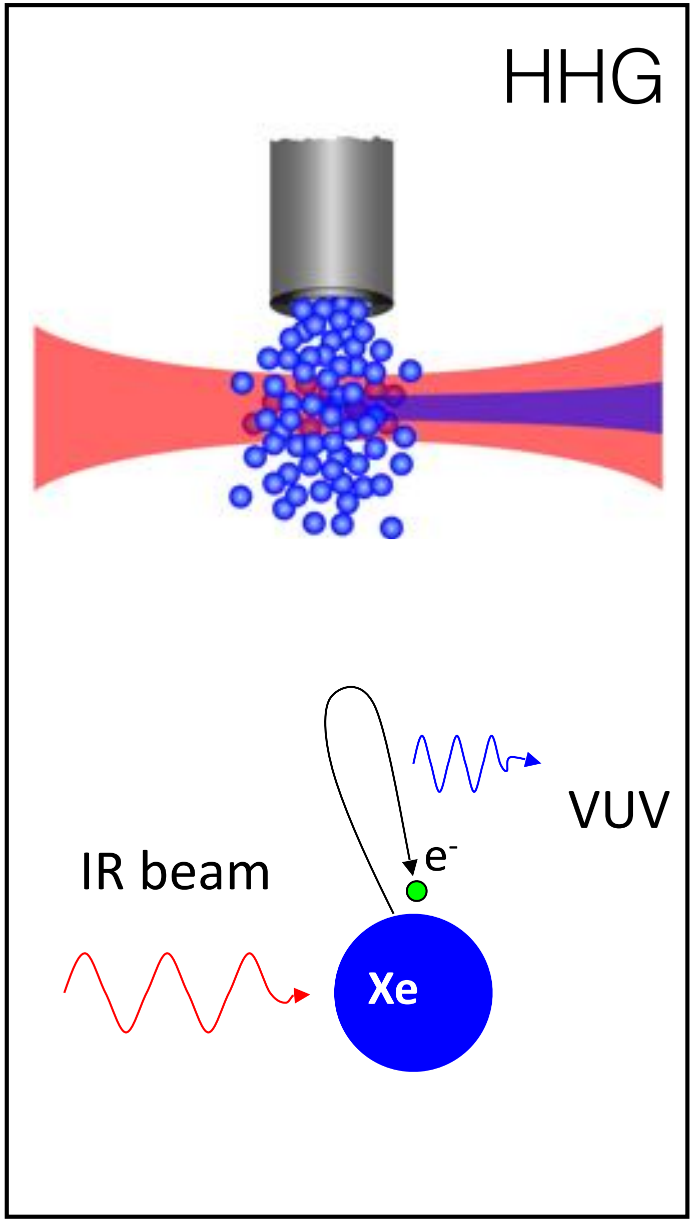
DLSR - diffraction limited storage rings
FEL - free electron lasers
HHG - high harmonic generation sources

coherent photon flux $\propto$ source brightness * wavelength^2

Coherent soft/hard x-ray sources

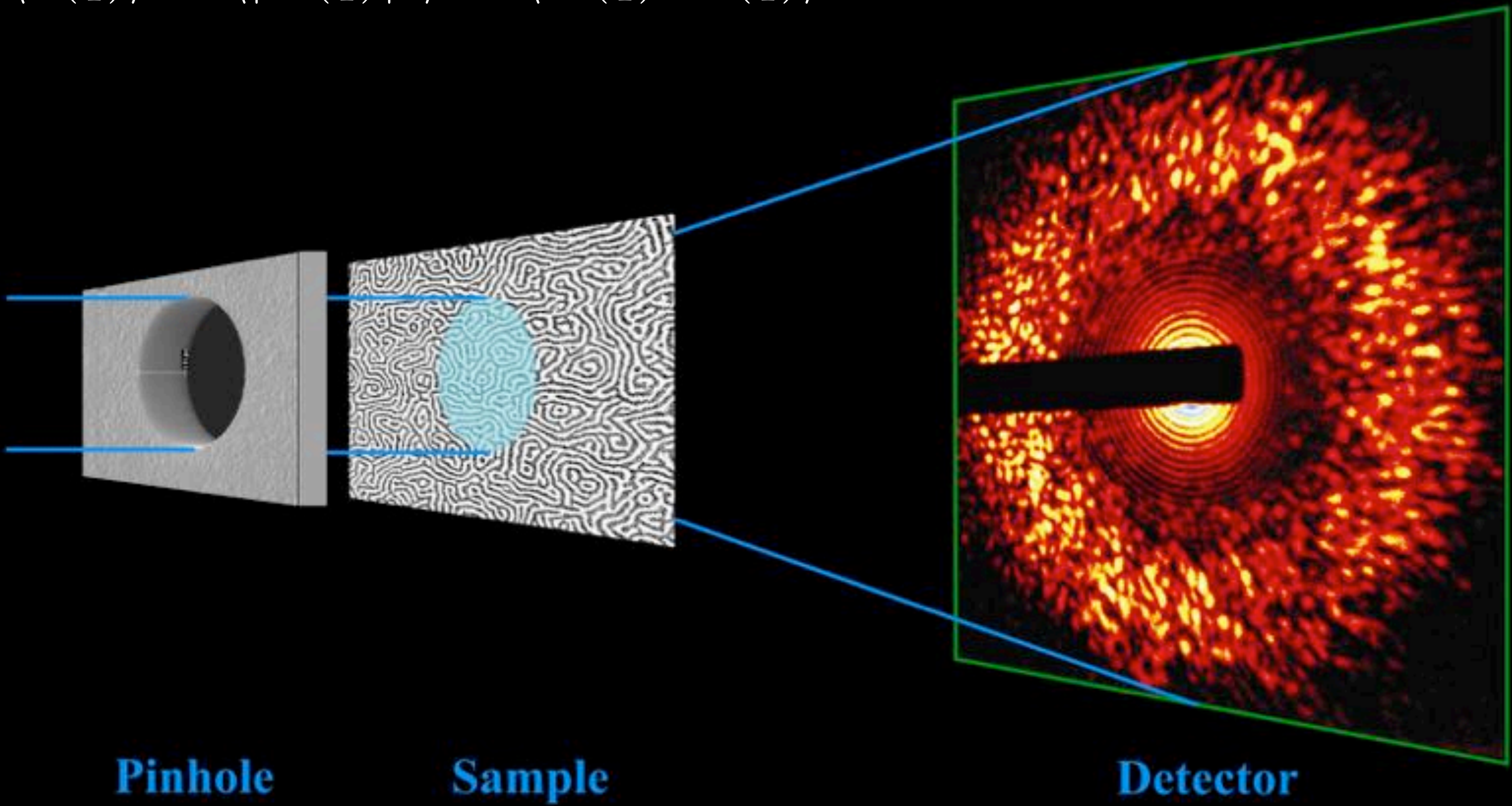






Scattering and Phase Problem

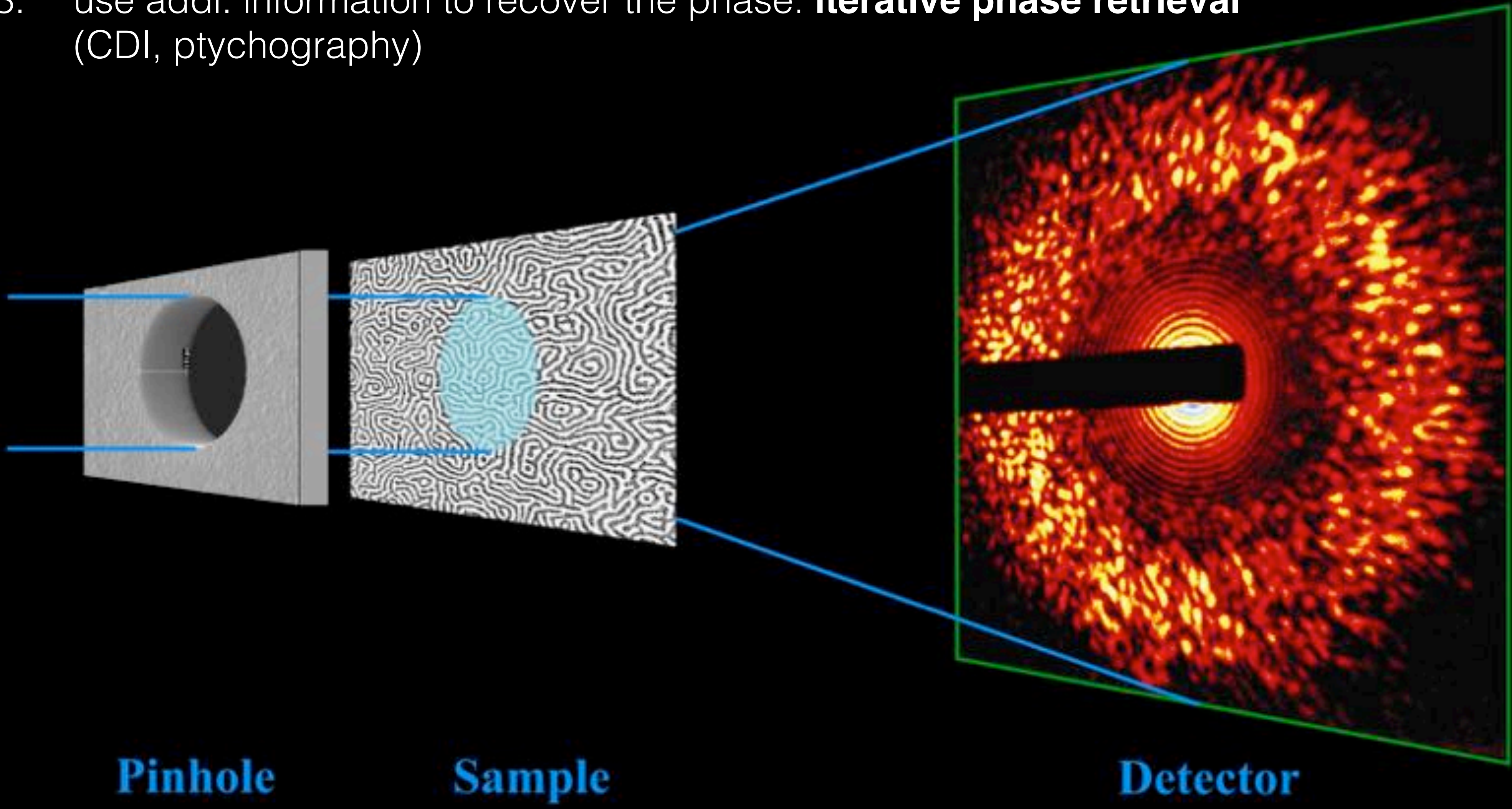
$$\langle I(q) \rangle = \langle |E(q)|^2 \rangle = \langle E(q) E^*(q) \rangle$$



Scattering and Phase Problem

Solutions:

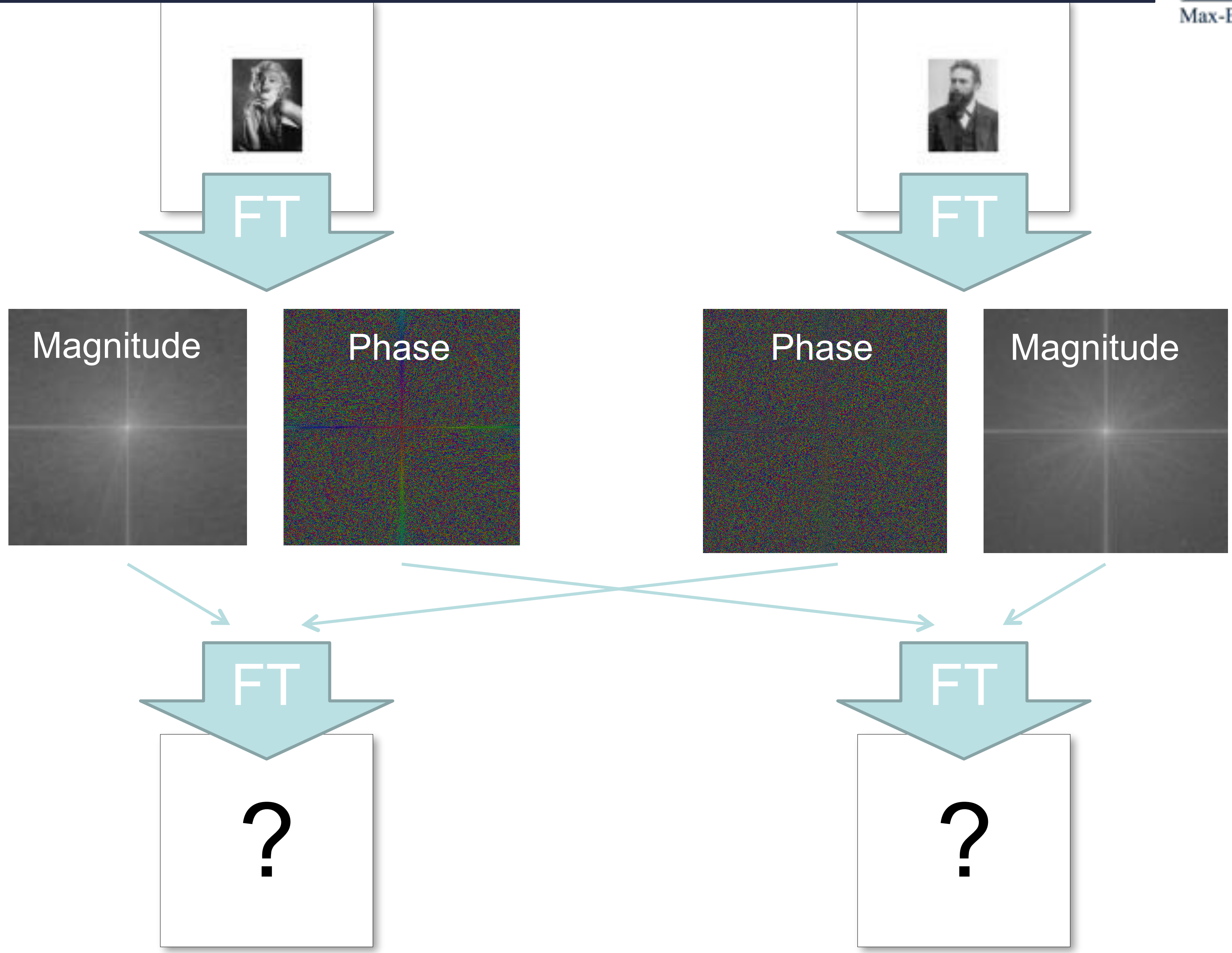
1. use a **lens** to form an image
2. encode the phase: **holography**
3. use addl. information to recover the phase: **iterative phase retrieval** (CDI, ptychography)



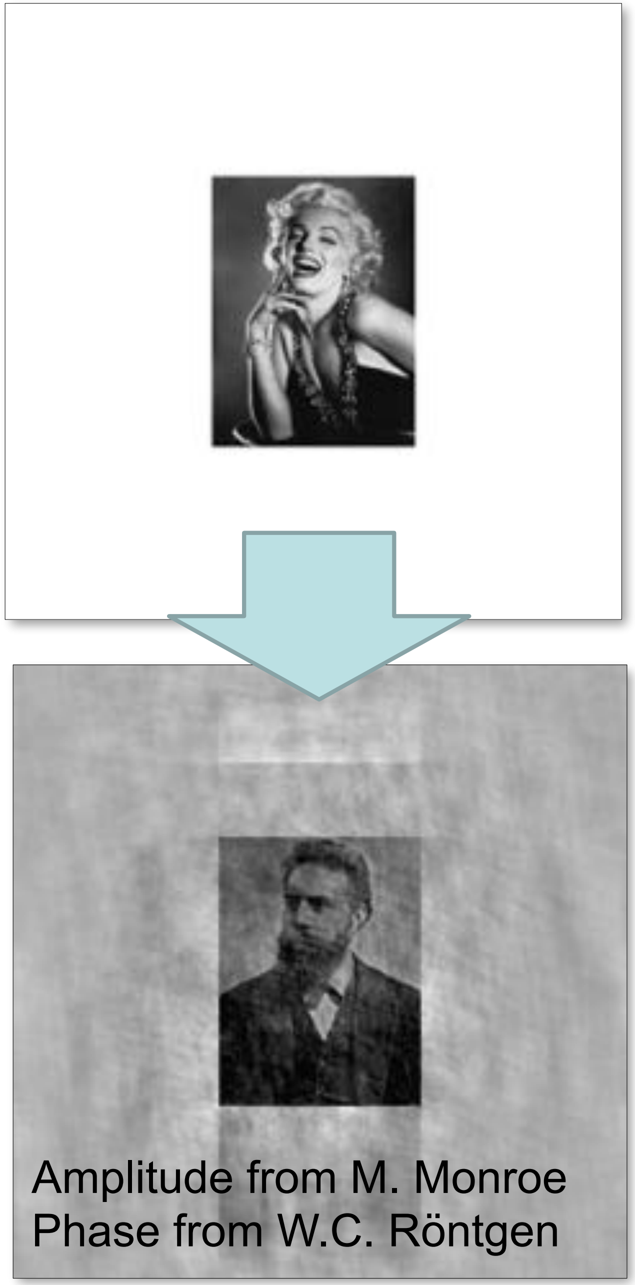
Why care about the phase?

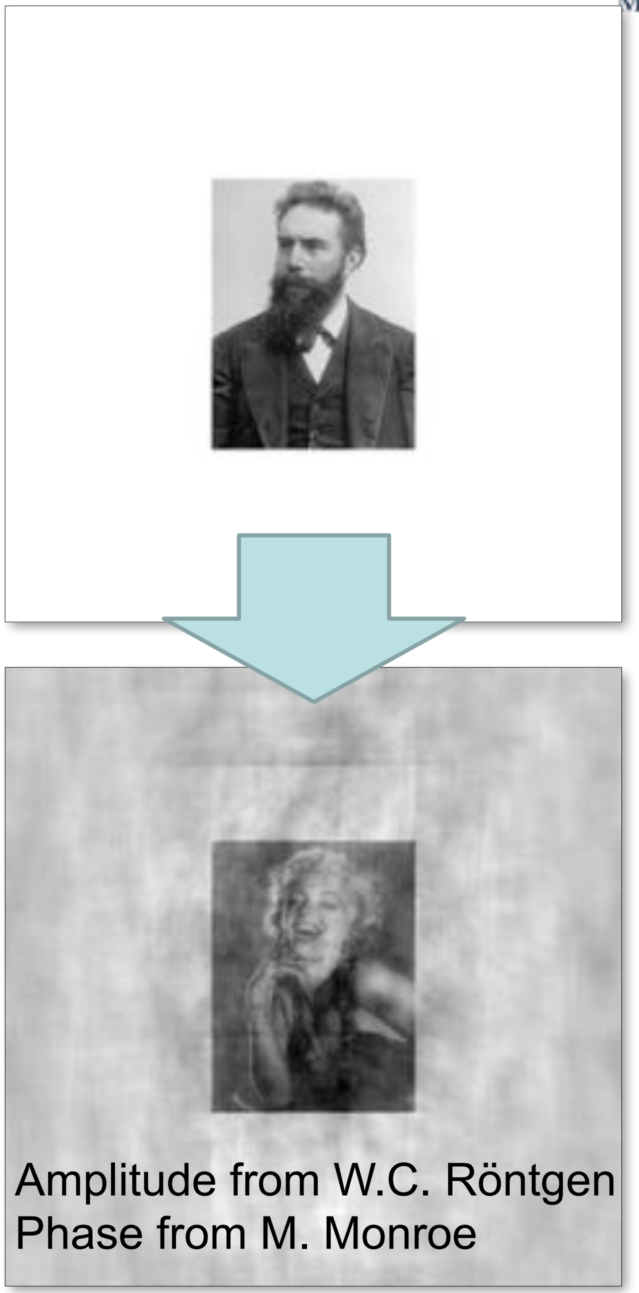
Pure absorption objects — No phase shift

A computer experiment...



Phase carries crucial information !





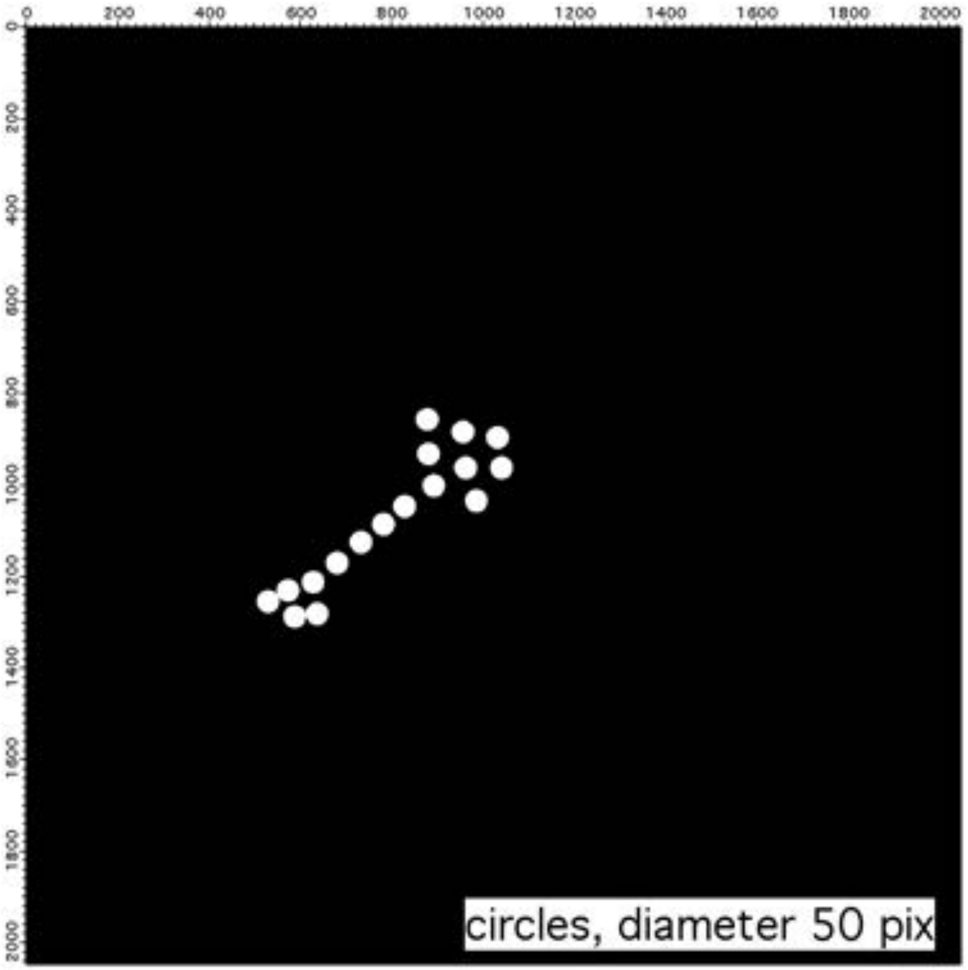
$I(q) = |E|^2 = EE^*$

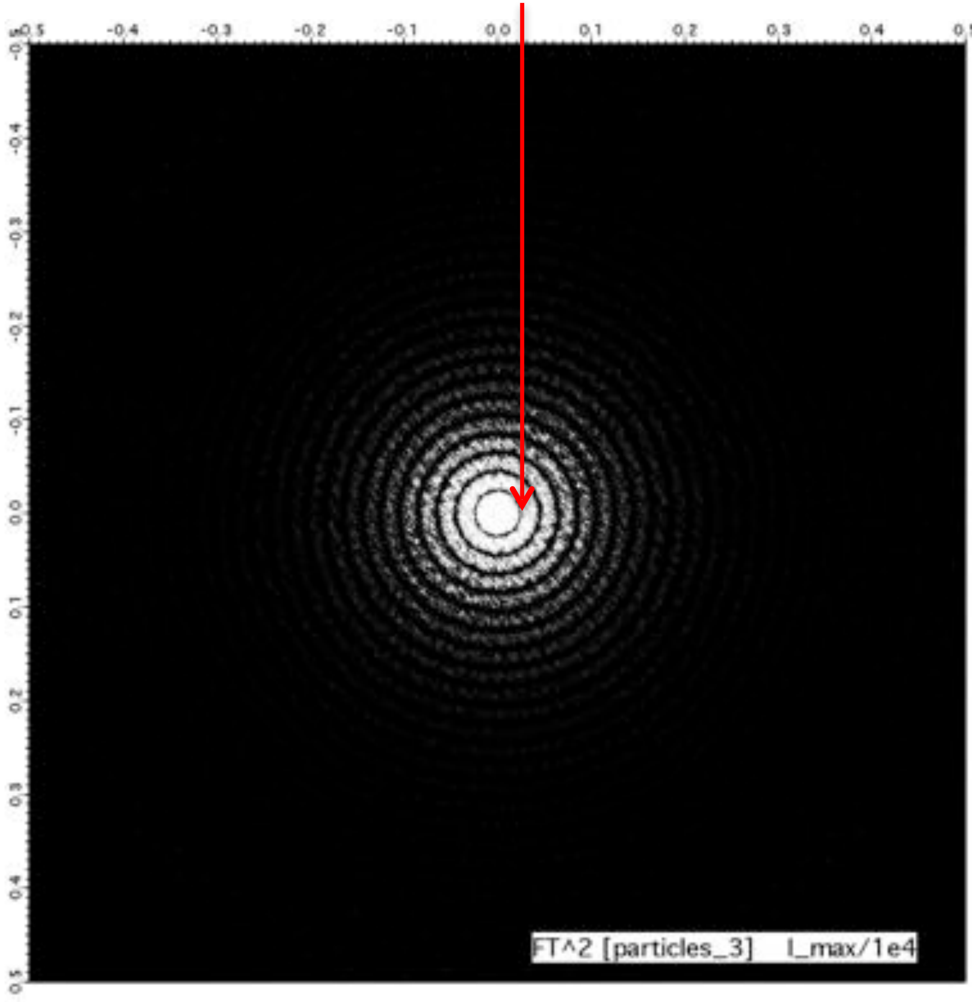
Let's see what can be done without knowing the phase

1. Minimum circ. aperture:

$$I(r) = I_0 \cdot \left(\frac{J_1(2\pi r)}{\pi r} \right)^2$$

J_1 Besselfkt (1)





The diffracted intensity is not easily translated into an image of the object

$\sin \alpha = 0.61\ (\lambda/r)$

depends on shape of aperture

Cnvolution theorem of the Fourier transform

$$\mathcal{F}[f * g] = \mathcal{F}[f] \cdot \mathcal{F}[g]$$

Convolution

Fourier Transform of f

FT of g

$$f * g = \mathcal{F}^{-1}[\,\mathcal{F}[f] \cdot \mathcal{F}[g]\,]$$

Consequence

If e(x,y) is the transmission of a 2D sample (or “exit wave”), then FT{a} •FT*{a} is the intensity at the detector

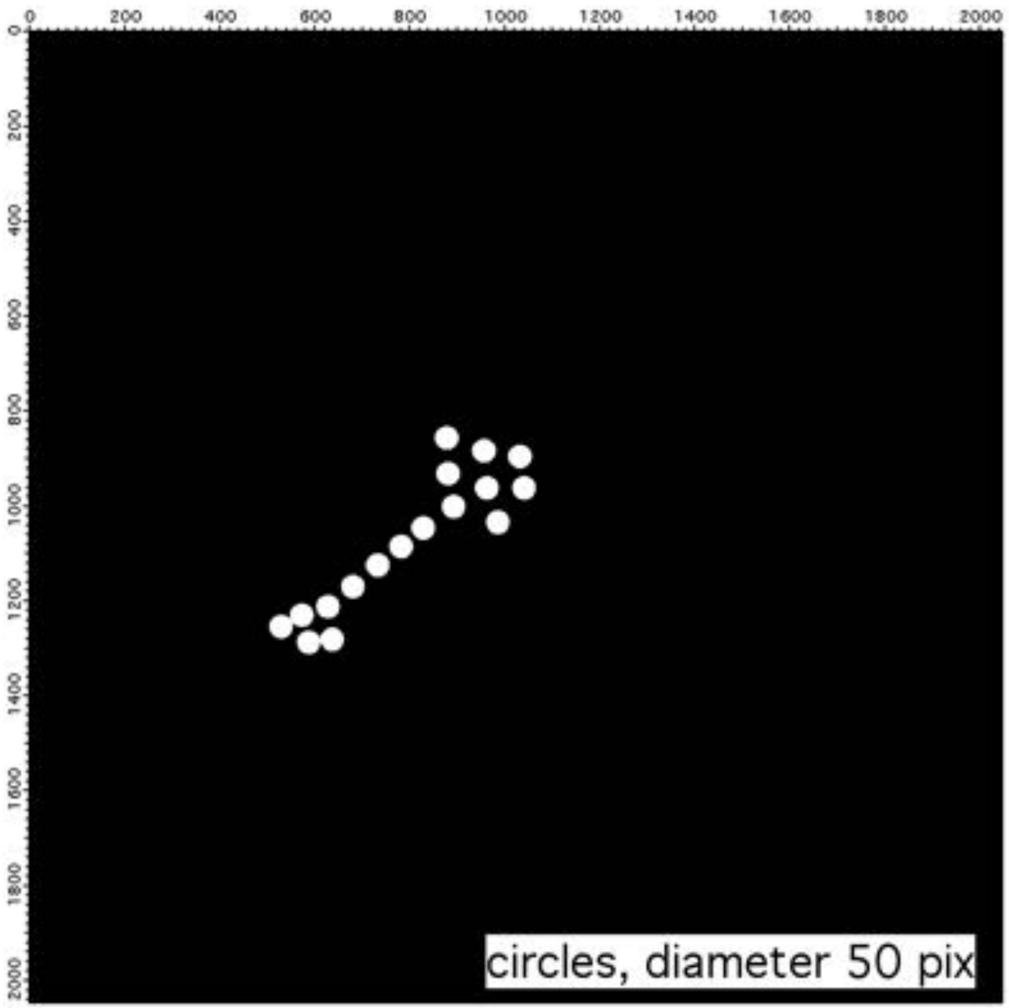
$$\mathcal{F}[\, e * e \,] = \mathcal{F}[e] \cdot \mathcal{F}[e]$$

$$e * e = \mathcal{F}^{-1}[\, \mathcal{F}[e] \cdot \mathcal{F}[e] \,]$$

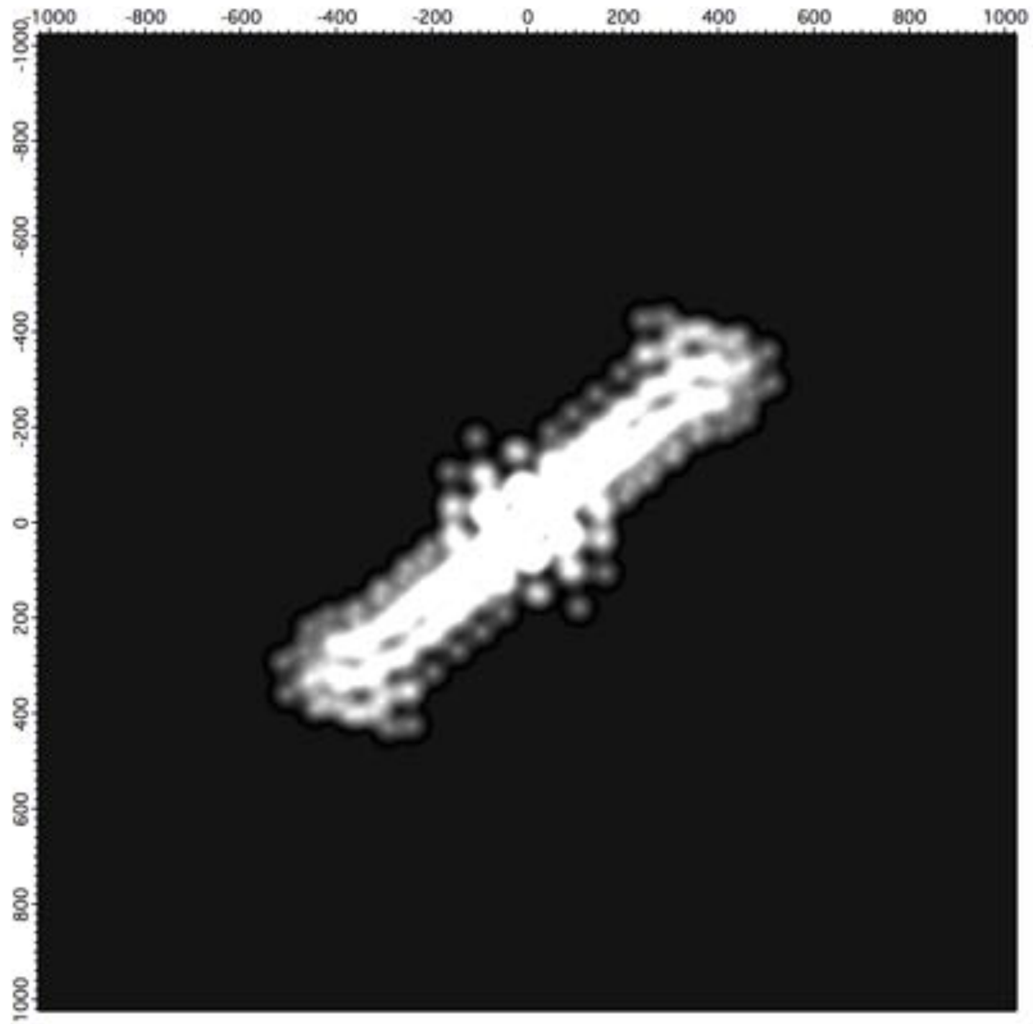
The inv. **Fourier transform of the detected intensity** is the spatial convolution of the real space object (e(x,y)) with itself. „**Patterson Map“**

Patterson Funktion

Objekt im Realraum

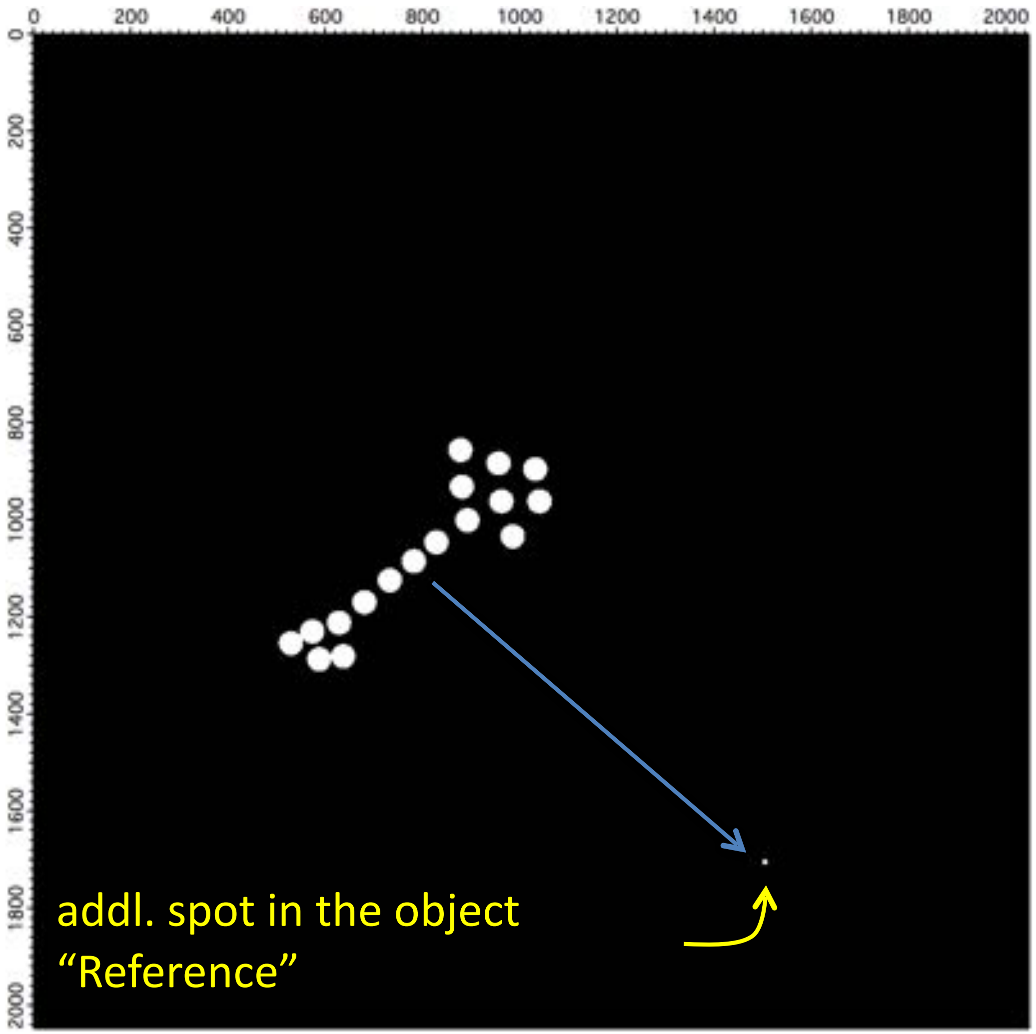


Patterson Map

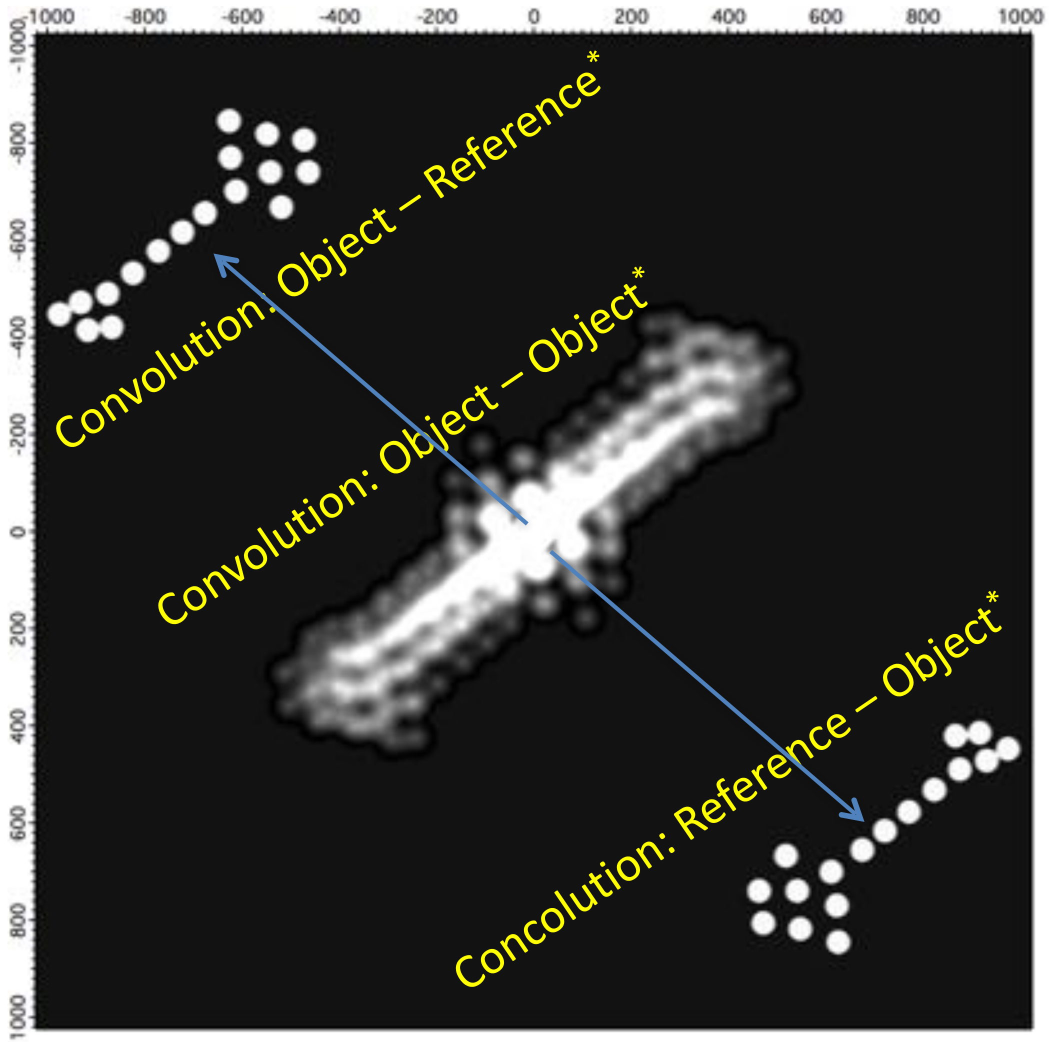


And now, an additional spot makes all the difference!

Object in real space



Patterson Map

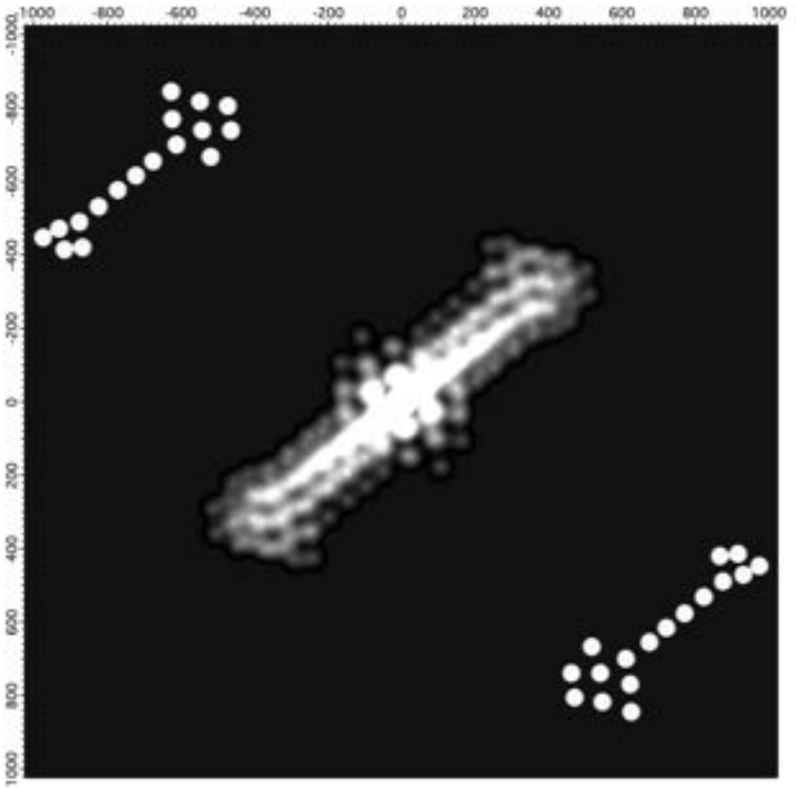


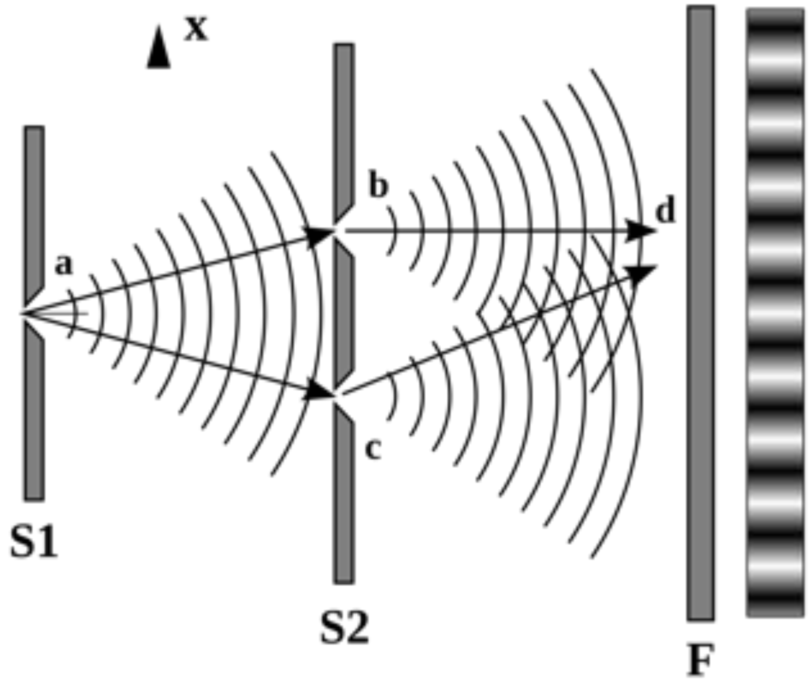
the reference aperture makes the Patterson Map easily interpretable: **Image** of the object !

Holography circumvents the phase problem

Why does this work?

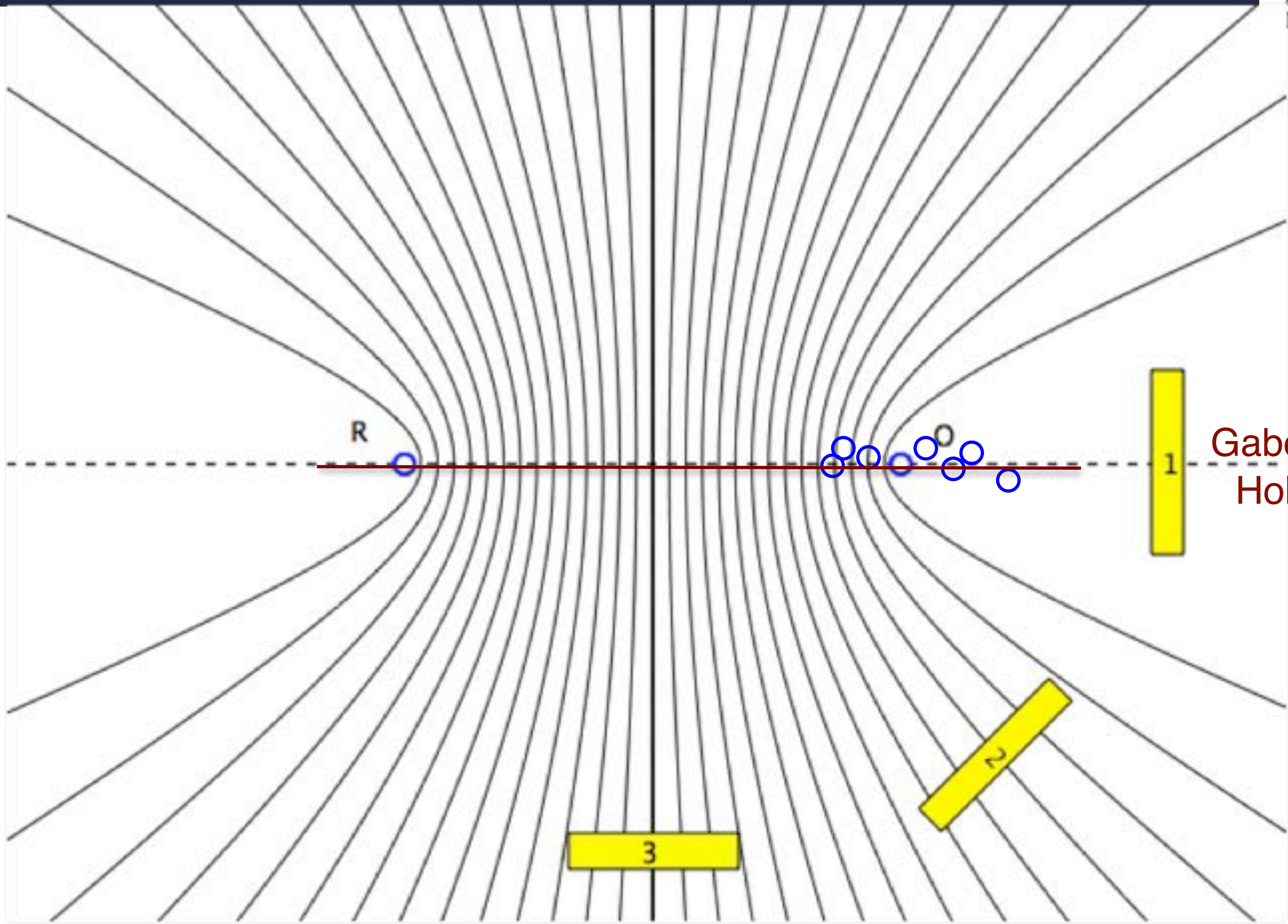
As in a double slit experiment, the relative **phase of the object wave is encoded** relative to a reference wave.





Wikipedia/Stannered

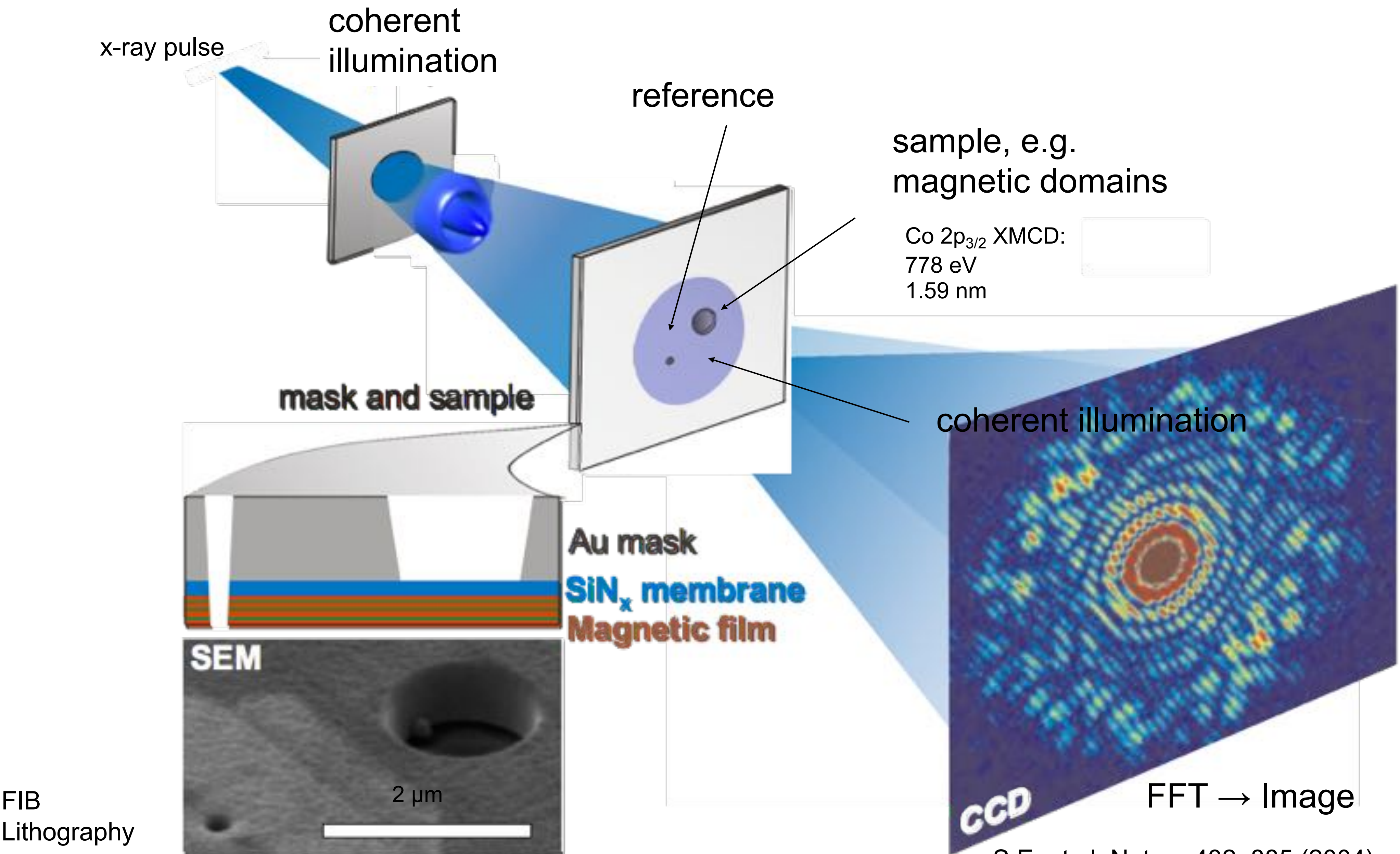
Holography Geometries



Gabor „in-line" Holography

Fourier Transform Holography

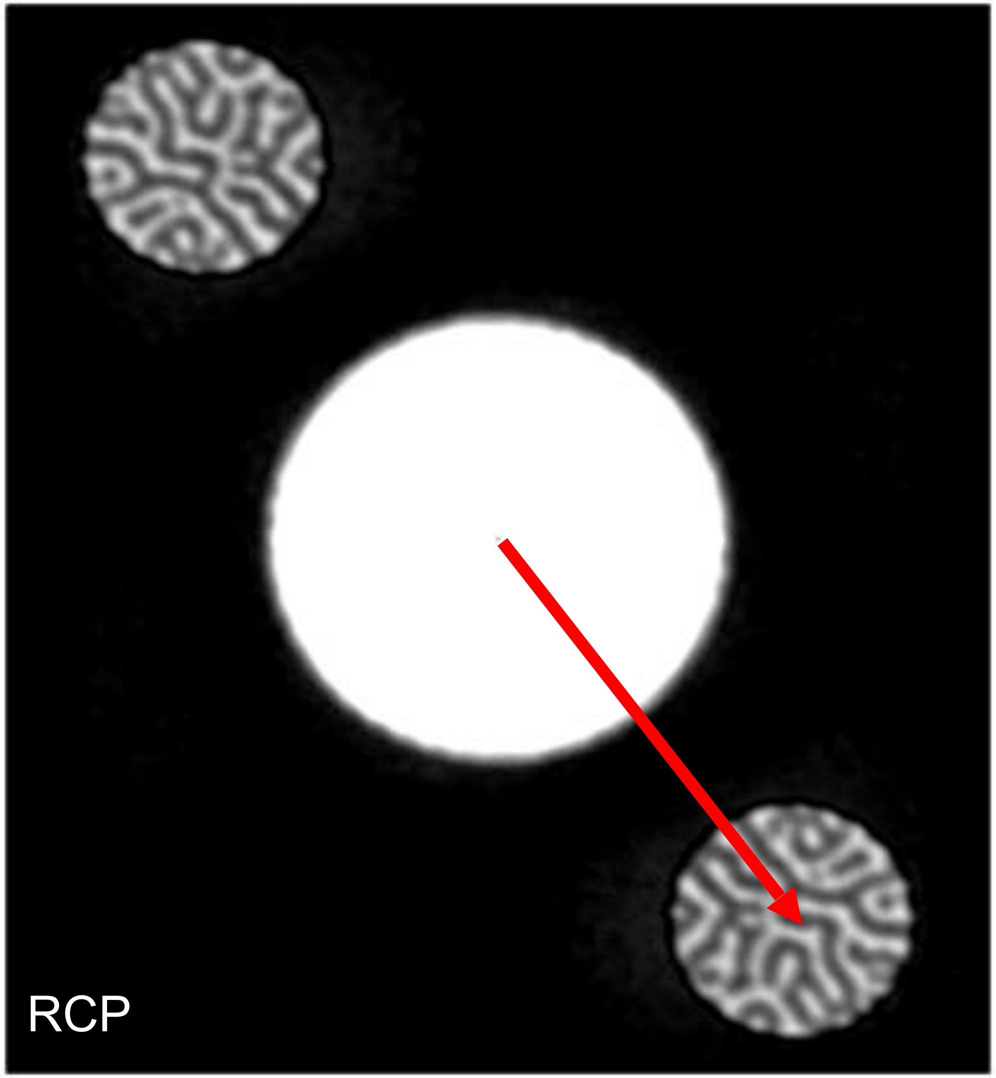
Mask-based x-ray Fourier transform holography



S.E. et al, Nature 432, 885 (2004)

Digital Image Reconstruction

FFT = Patterson Map

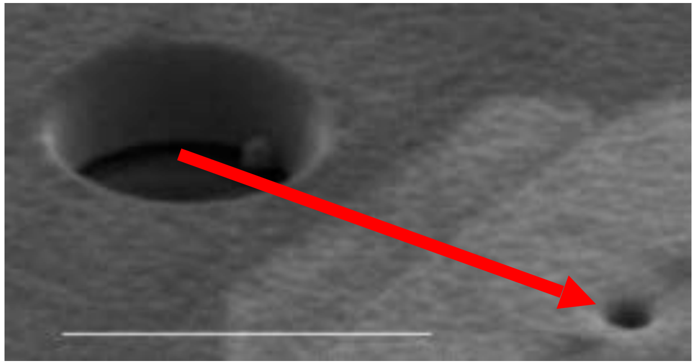


S.E. et al, Nature **432**, 885 (2004)

Convolution theorem applied to diffraction:
FT(diffraction) = Autocorrelation (Object)

FT (a ⊗ b) = FT(a) · FT(b)

$(a \otimes a) = FT^{-1}\{FT(a) \cdot FT(a)\}$



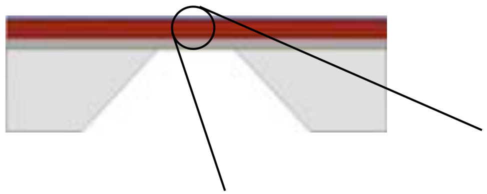
a (x,y) real space object

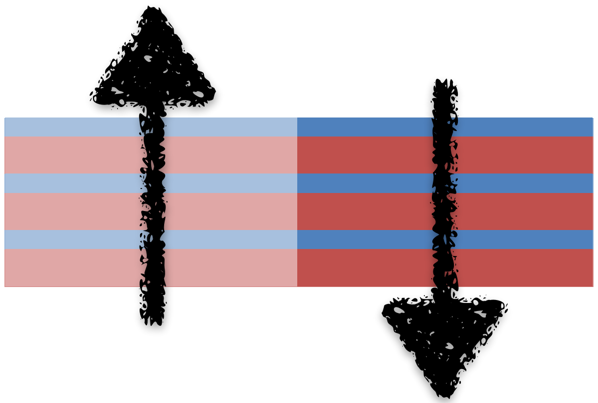
FT(a) · FT(a) diffraction intensity

- result = complex 2D matrix, containing magnitude and phase!
- spatial resolution today typically 30 - 50 nm
- Note: Hologram = oversampled diffraction pattern. Iterative refinement possible. See:
  S. Flewett, et al. PRL **108**, 223902 (2012)
  S. Flewett et al, Optics Express **28**, 29210 (2012)

A prototypical sample system: Ferromagnetic Domains
in ML with perpendicular magnetic anisotropy

Side view



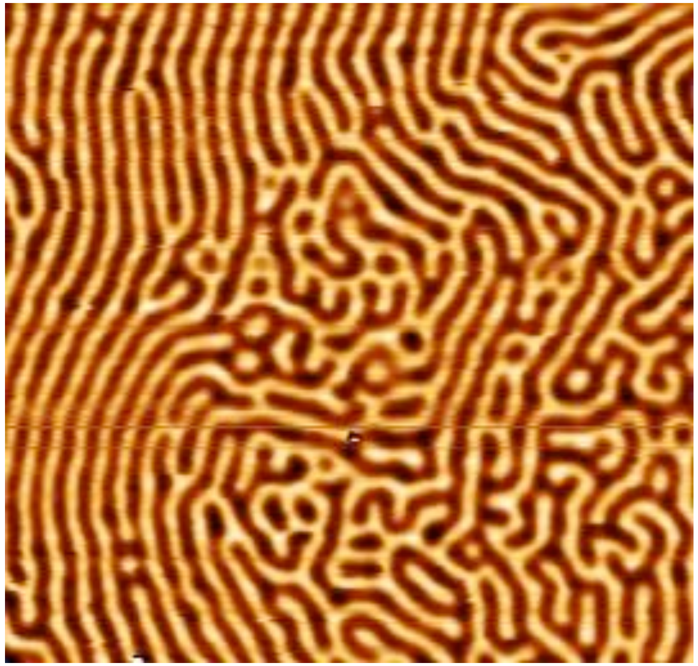


e.g.

SiN_x / Pd (20 Å) / [Co (4 Å) / Pd (2 Å)]$_{20}$ /Al (30 Å)

- perpendicular magnetic anisotropy
- Magnetron sputtering
- polycrystalline, grown on thin Si_3N_4 membrane

MFM, top view



ca. 5 μm x 5 μm

Contrast mechanism:

X-ray Magnetic Circular Dichroism

XMCD

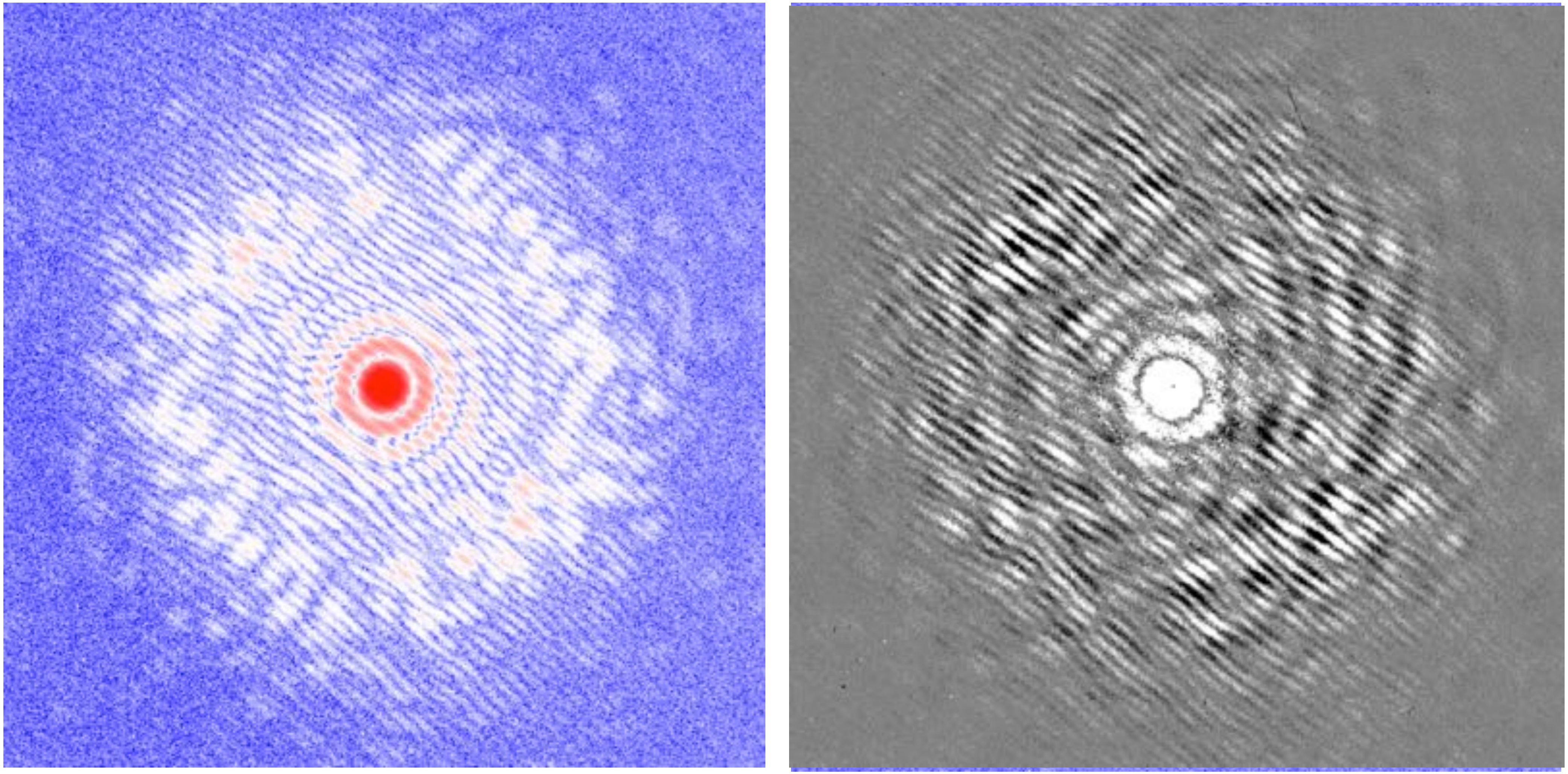
(i.e. resonant scattering, need suitable photon energy)

Dichroic Hologram (XMCD contrast)

CCD Detector Image

Right circular polarized

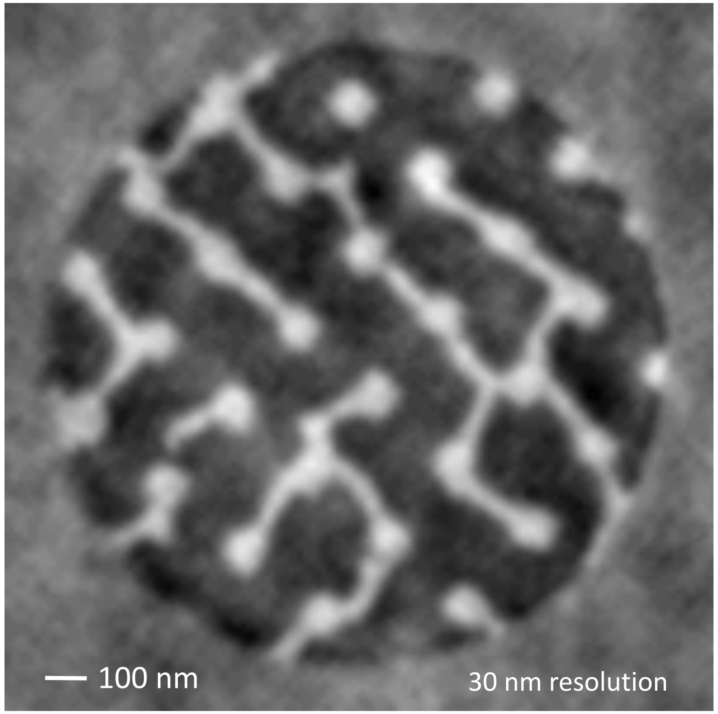
Difference (RCP – LCP)



log z-scale

Ferromagnetic Domains in PMA Multilayers.
L vs. M-edge.

∅ 1500 nm

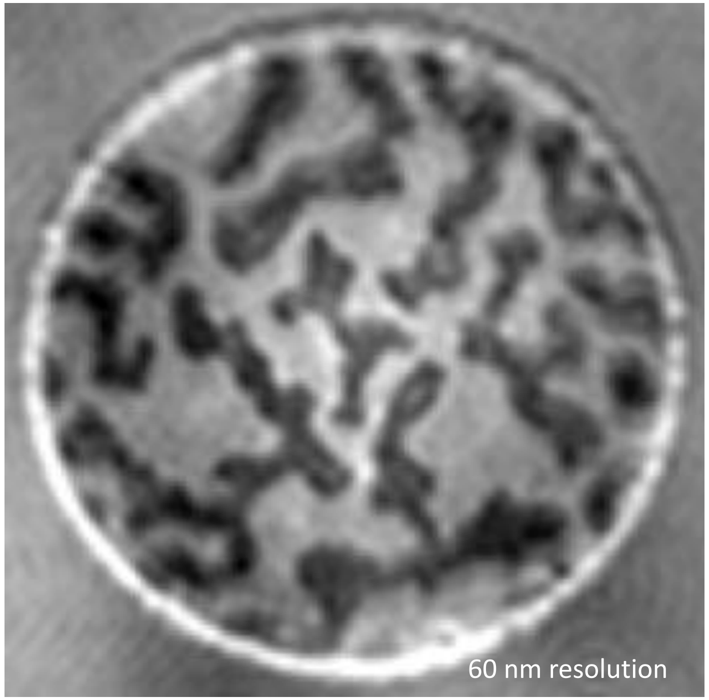


B. Pfau et al, *APL* ***99,*** 062502 (2011)

Co L_3 XMCD contrast

778 eV, λ = 1.6 nm

∅ 2000 nm



S. Schaffert, New J. Phys. **15**, 093042 (2013)

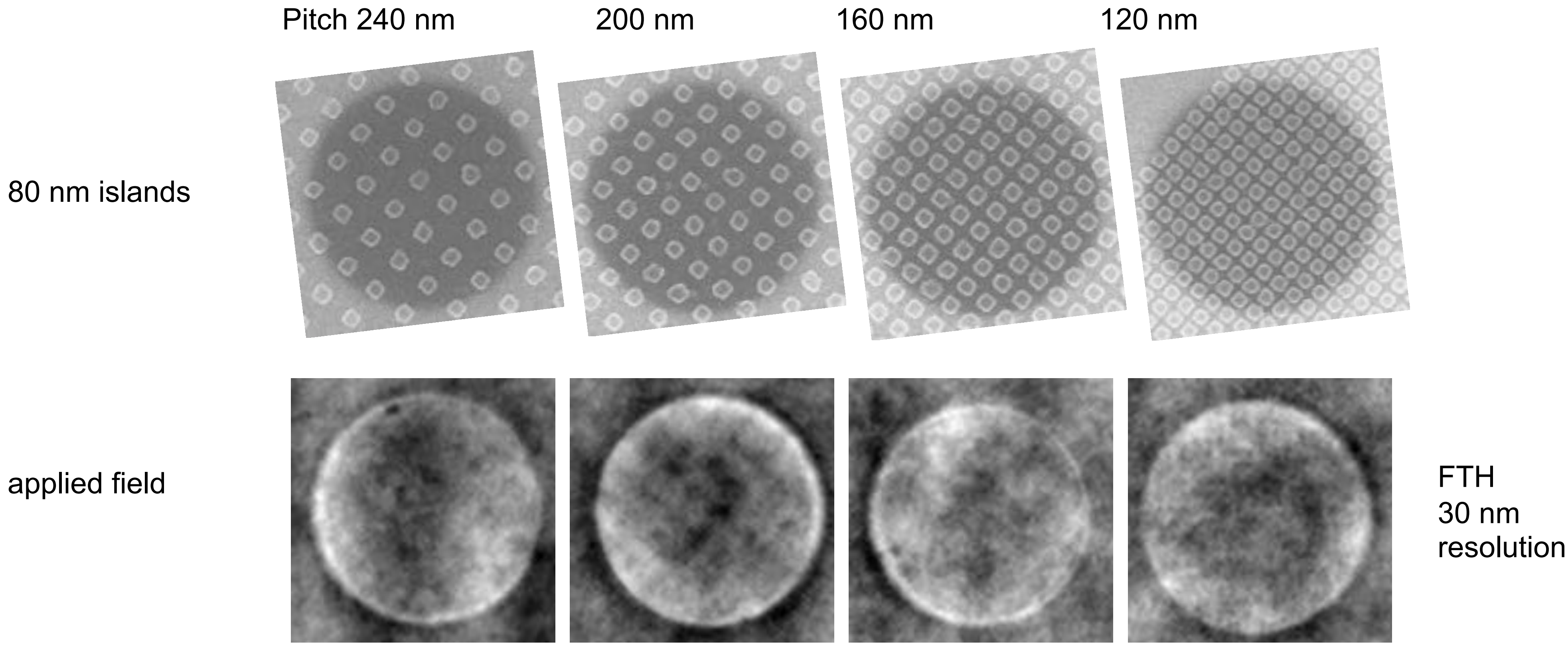
Co M_3 XMCD contrast

60 eV, λ = 20.8 nm

- we obtain amplitude and phase via a direct Fourier inversion of the hologram
- With very intense coherent x-ray pulses, *single shot images* are possible

T. Wang et al., Phys. Rev. Lett. **108**, 267403 (2012)

Switching behavior of magnetic nanostructures.
Data storage media (HGST BPM test)



map of the local magnetization
contrast: XMCD, helicity difference to suppress non-magnetic signal

B. Pfau et al, APL **99,** 062502 (2011) & APL **105**, 132407 (2014)

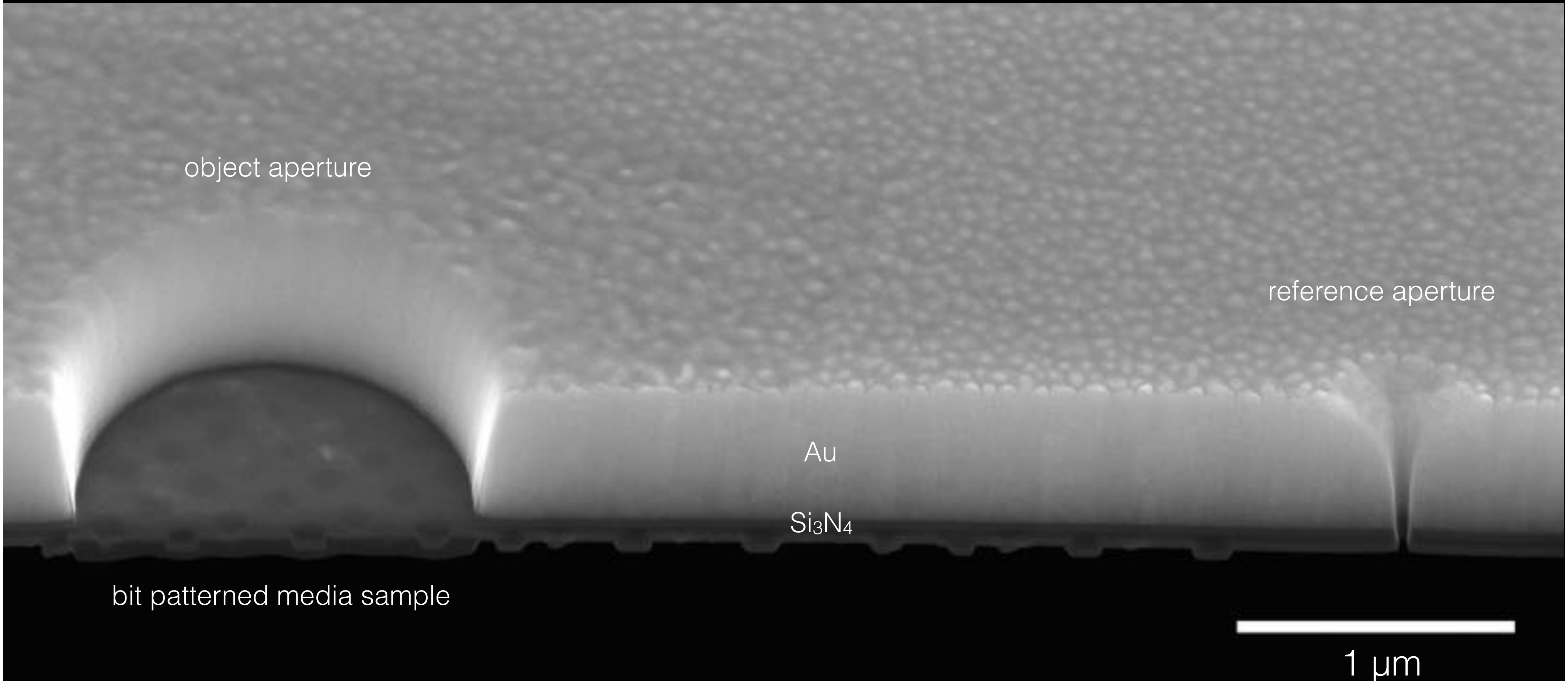
Long-term cooperation with O. Hellwig, HGST, now U. Chemnitz

Now we have the basics covered:

- FT Holography = coherent scattering with a suitable reference within the object plane.
- Off-axis geometry & far field → get image via direct Fourier Inversion.
- The complex wave field is encoded, i.e. amplitude and phase are accessible.
- Using x-rays, high resolution imaging is possible.
- q_{max} and ∅ reference limit resolution, state of the art ≈15 nm

Now, some extras:

FTH Mask for use with soft x-rays (≈1 keV)

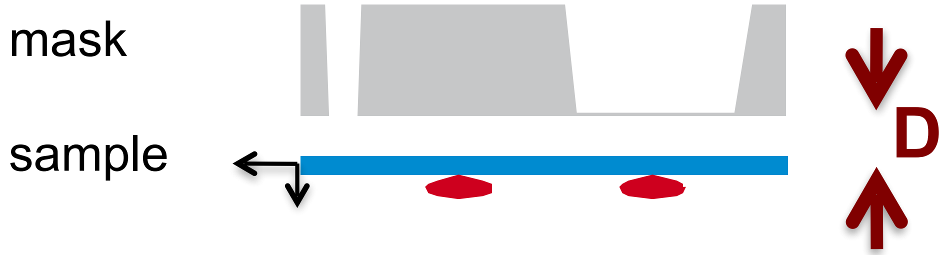


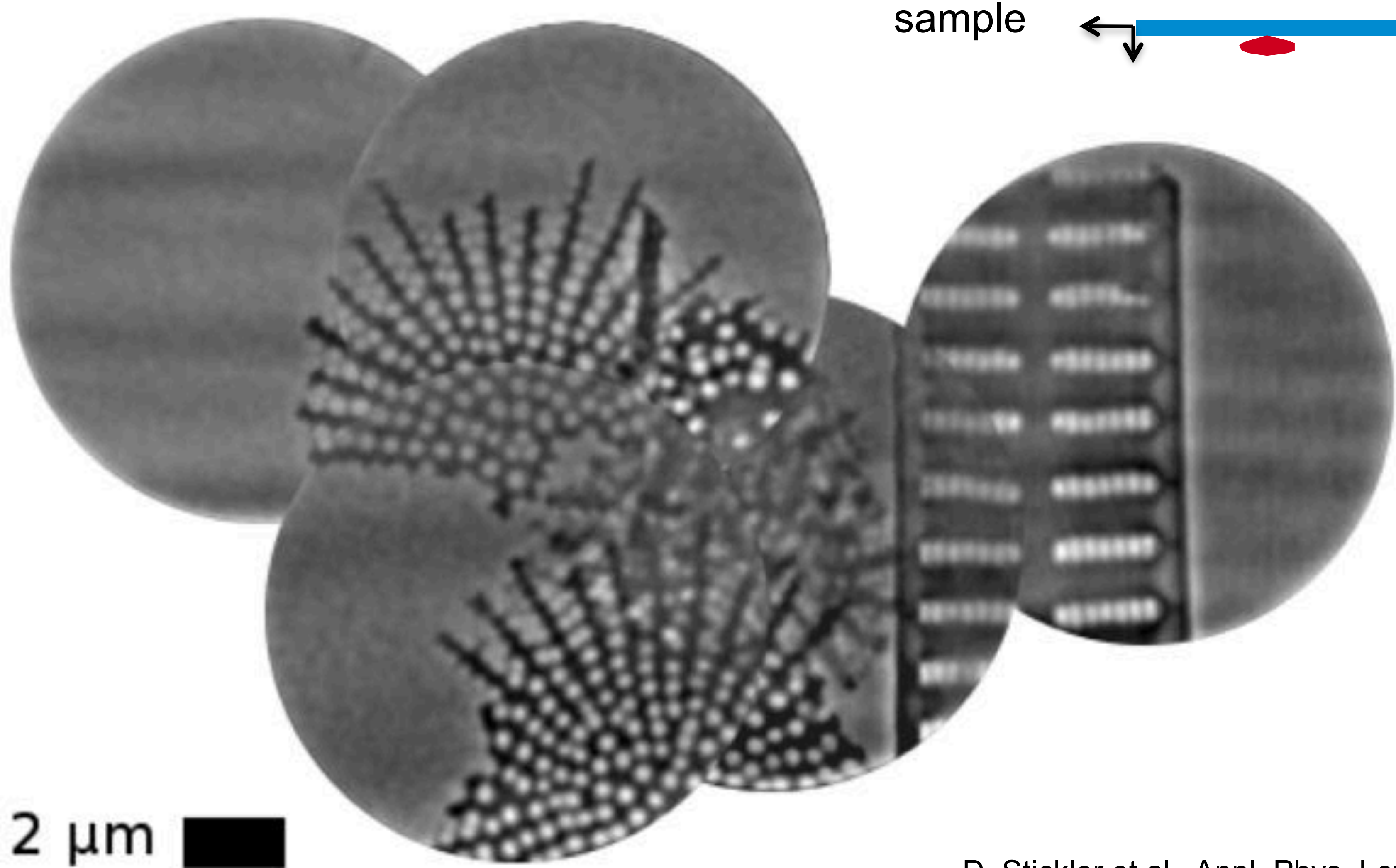
- generated via Focussed Ion Beam (FIB) lithography
- mask approach gives very clean data, but difficult to do for harder x-rays

C. Günther

Scanning Field of View / Backpropagation

Test Objects: Diatoms





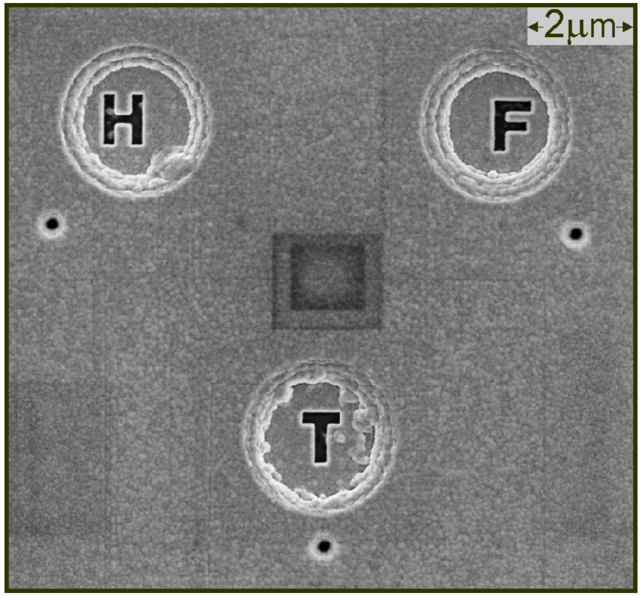
D. Stickler et al., Appl. Phys. Lett. **96** (2010)
this work: E. Gührs et al., Opt Express **17**, 6710 (2010)

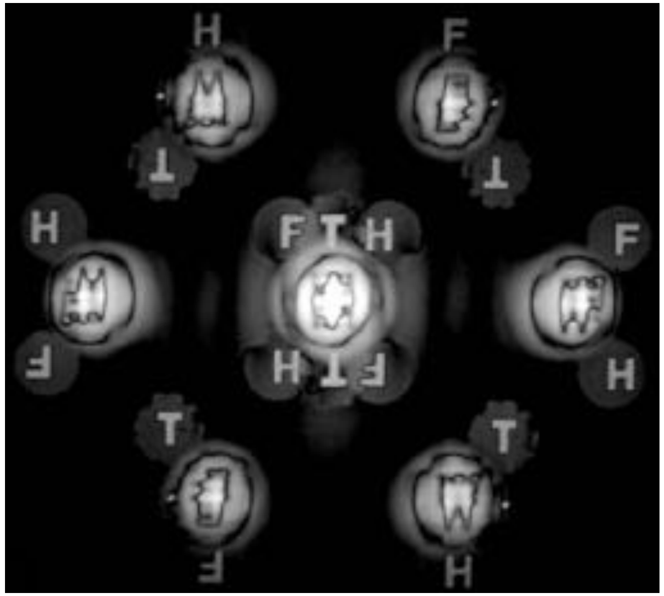
Object multiplexing

Multiple Objects

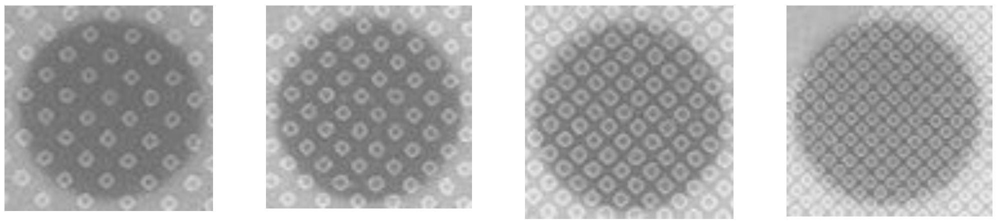
Each object is imaged by its own reference

Schlotter et al.,
Opt. Lett. **32**, 3110 (2007)

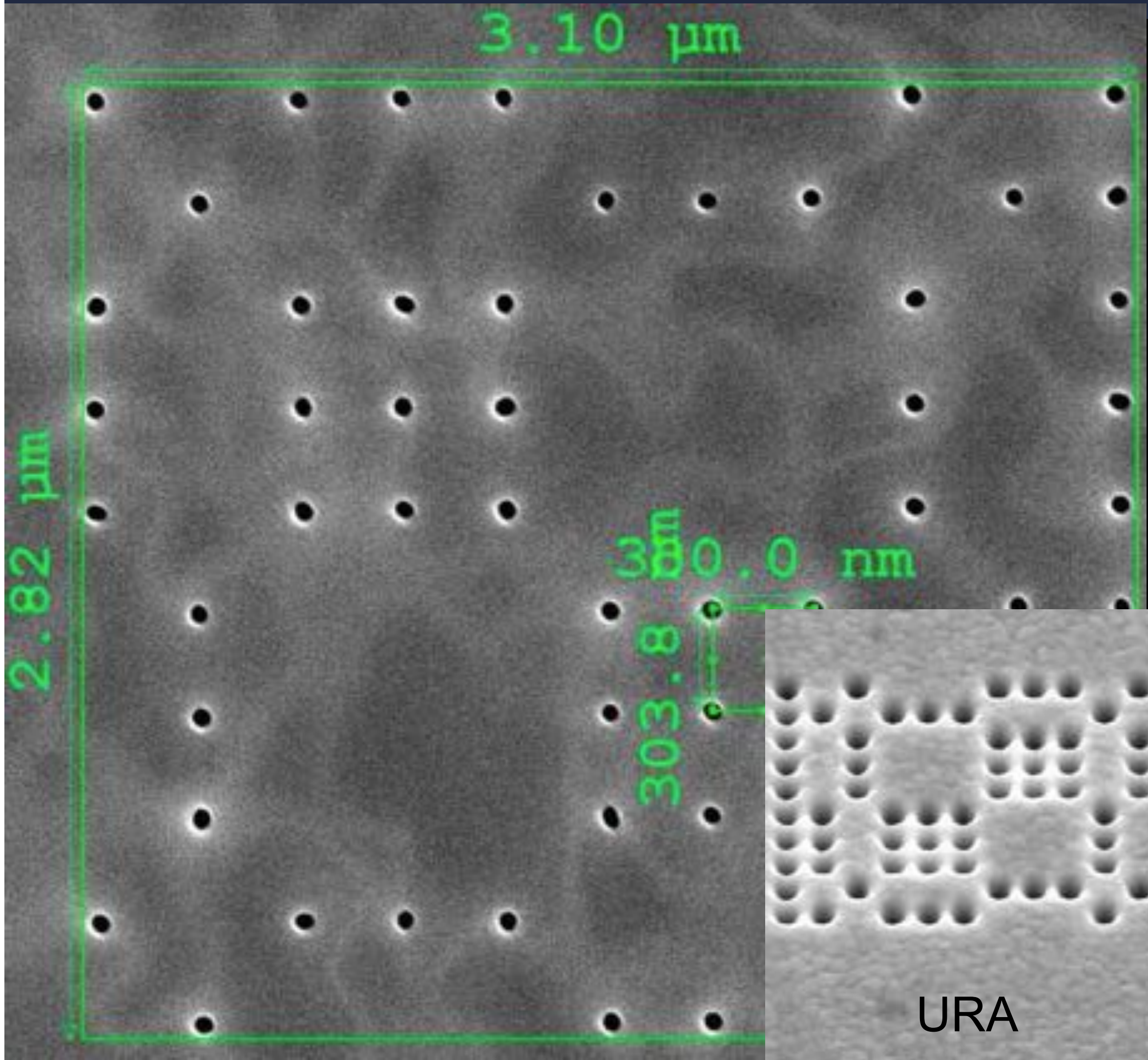


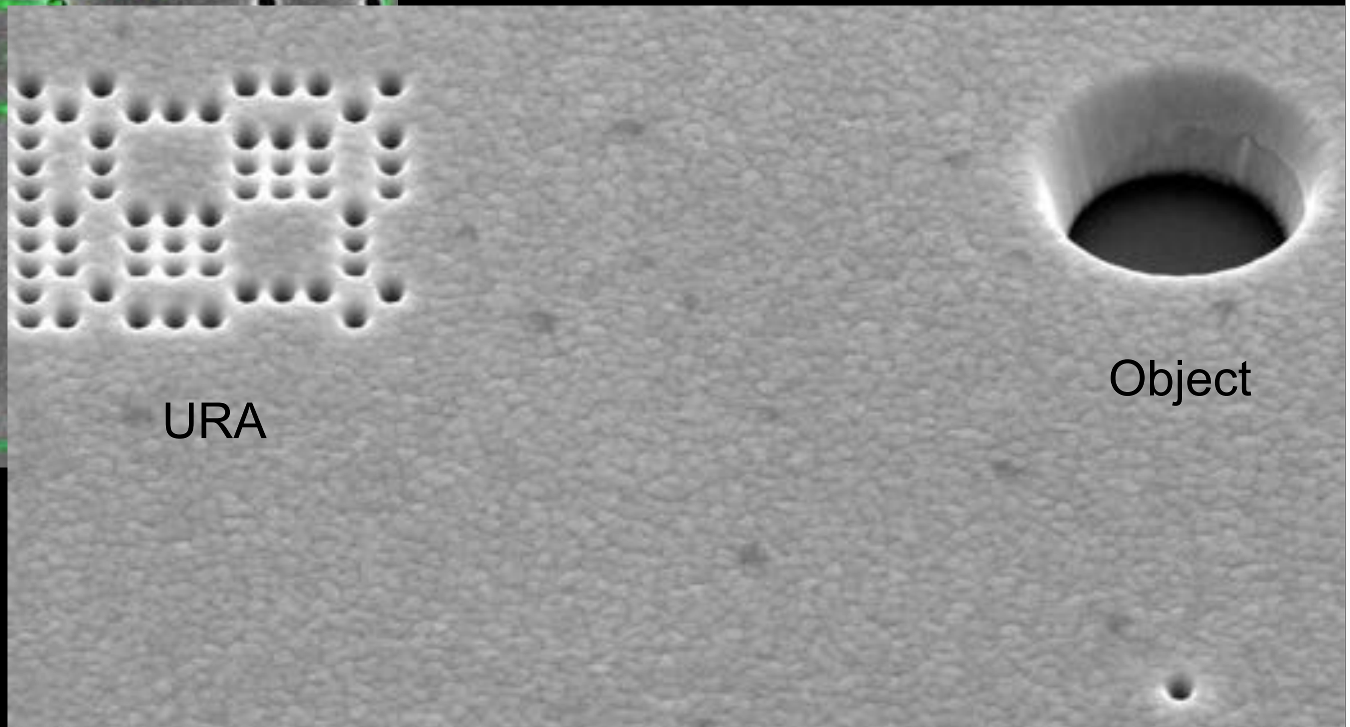


Concept was used in earlier Bit Patterned Media example.
Samples only few μm apart, all see same sample environment (B-field)



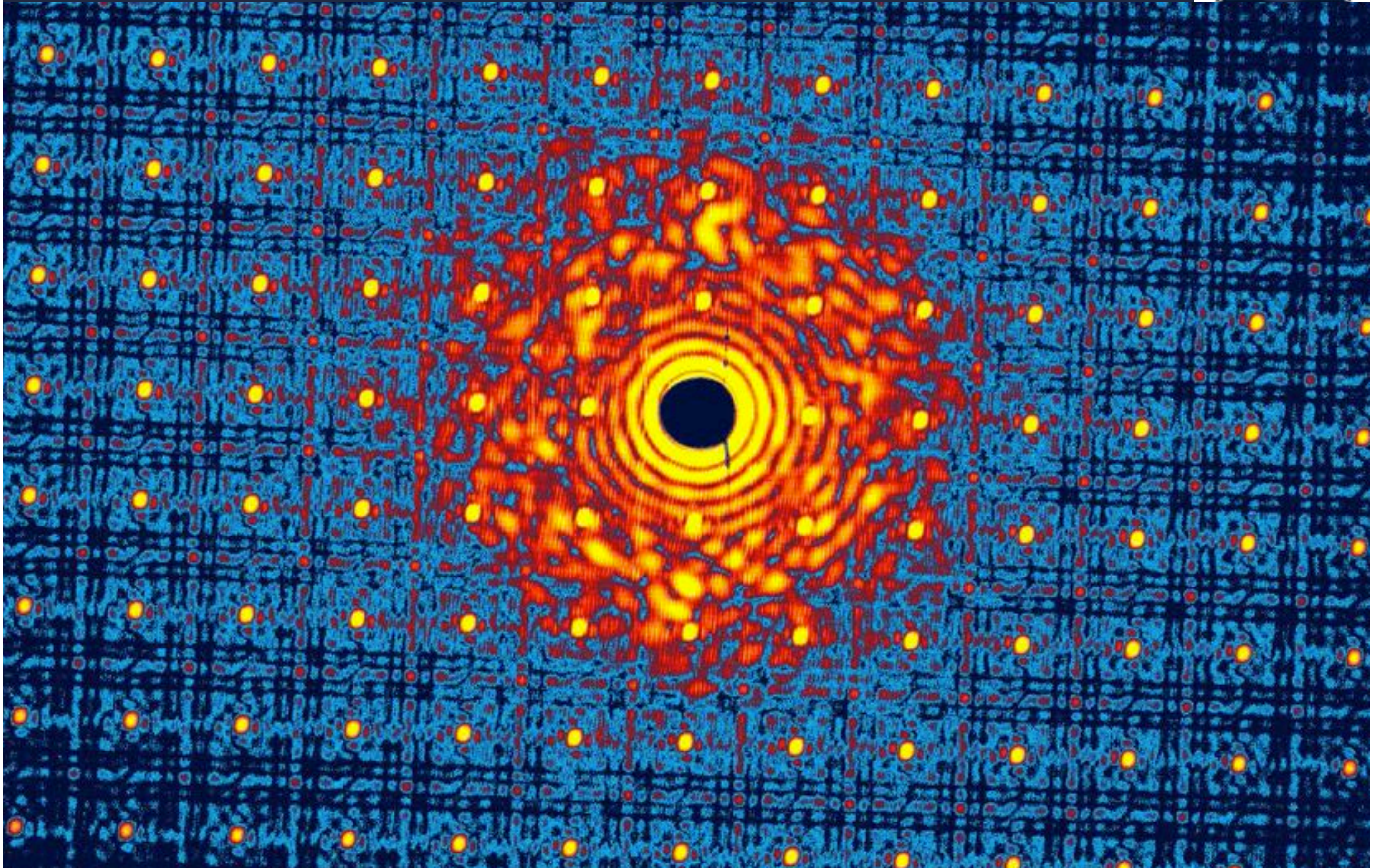
Many references: modified Uniformly Redundant Arrays





see also
S. Marchesini Nat. Phot. 2018

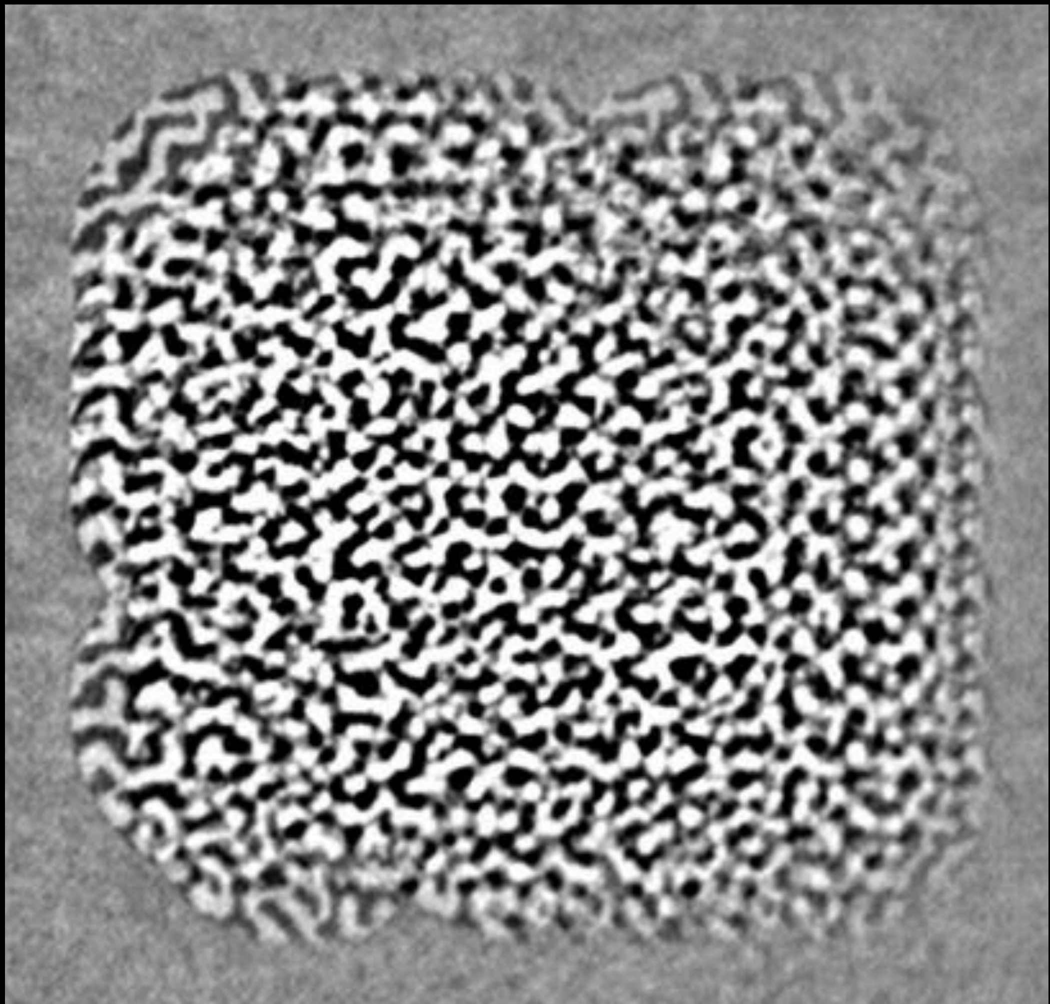
FT Hologram

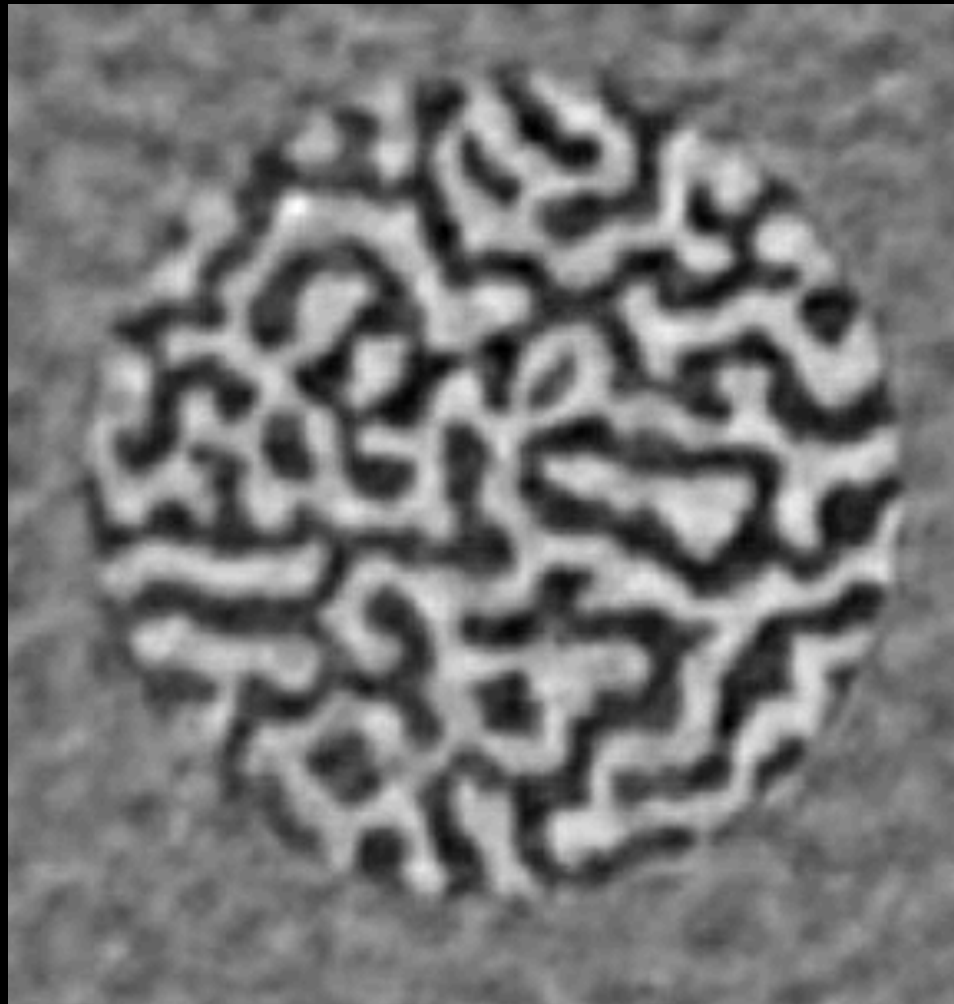


Coded aperture imaging

2D FFT

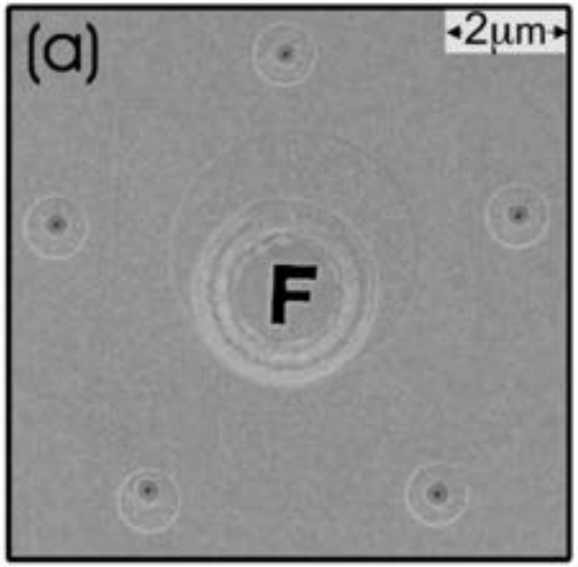
decode using URA Info (non-iterative):

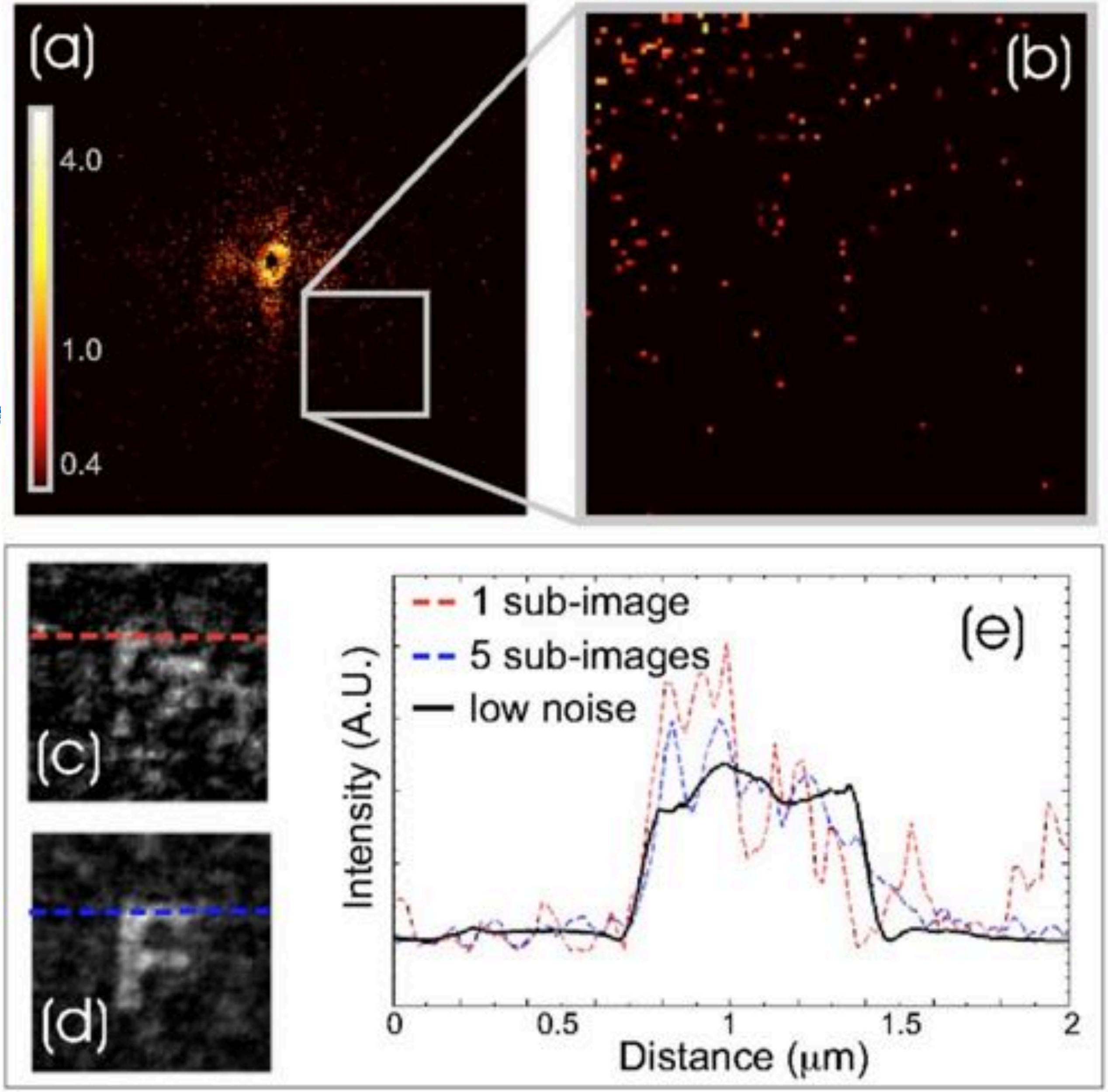




C. Günther, J.Opt. **19**, 064002 (2017)

FTH: robust reconstruction as phase is encoded



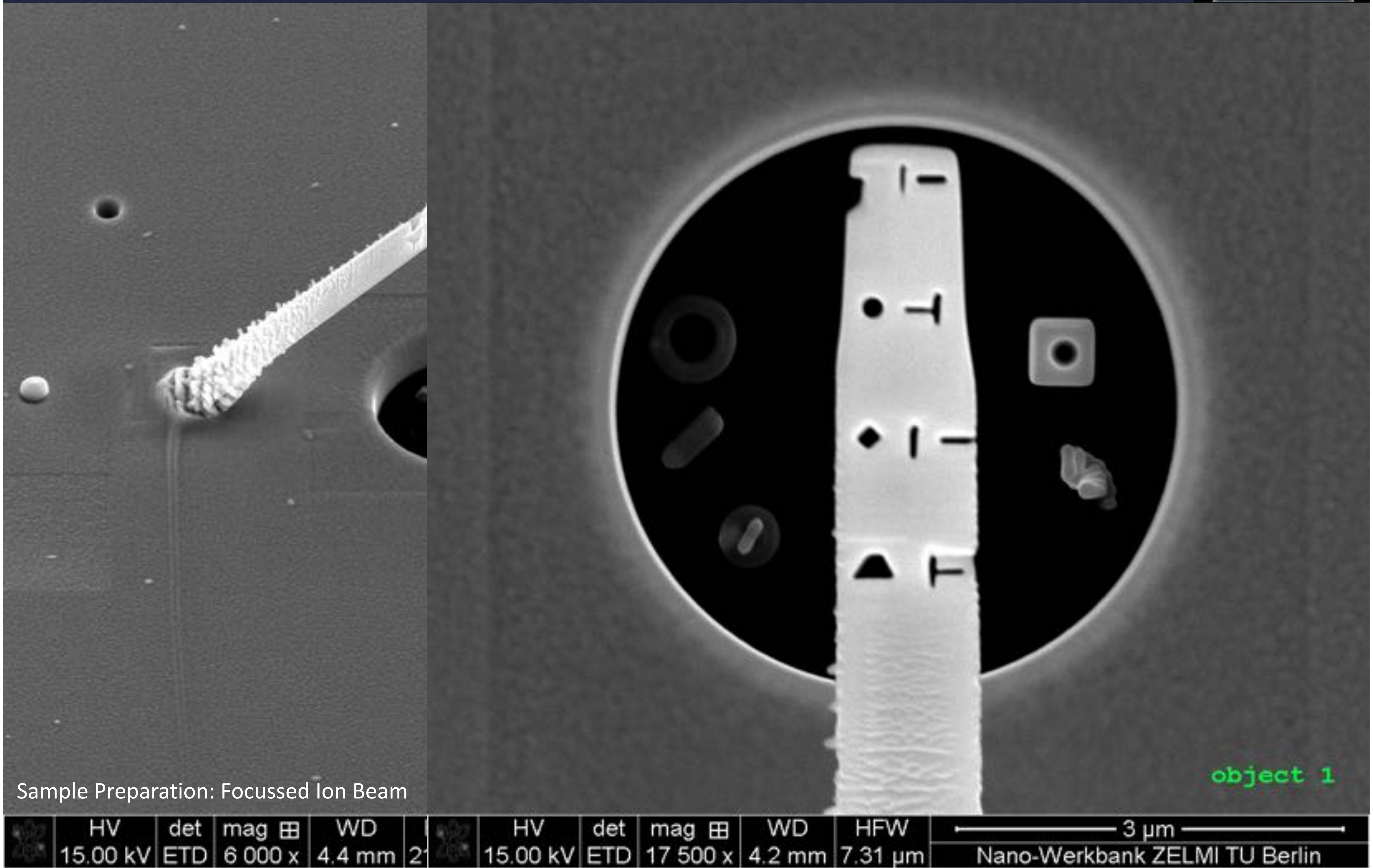


photons per pixel on CCD

W.F. Schlotter et al. , APL **89**, 163112 (2006)

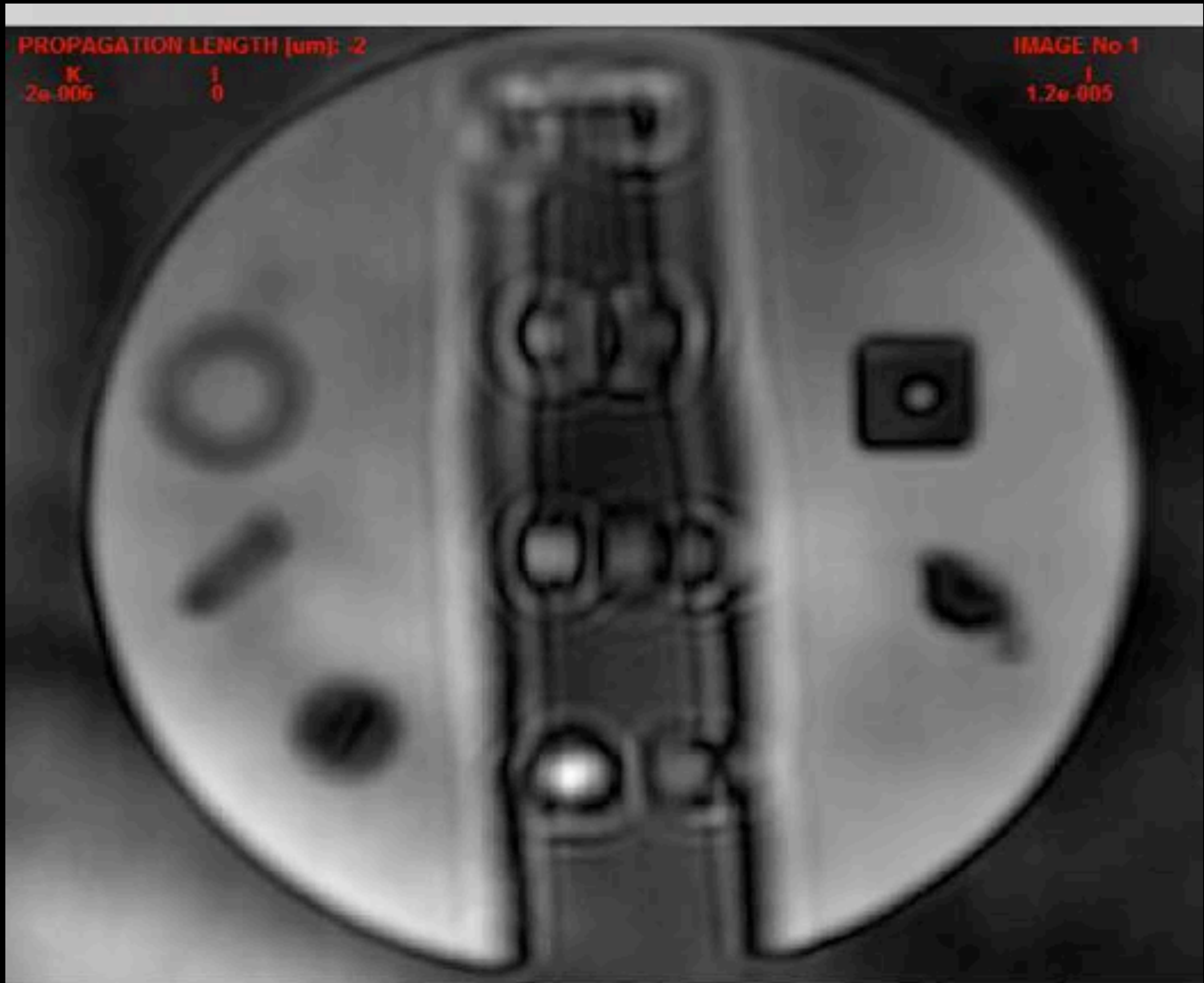
X-Ray Holography: Amplitude & Phase !

→ depth information possible



Sample Preparation: Focussed Ion Beam

3D in a single shot (high NA detection)



J. Geilhufe et al., OpEx **22**, 24959 (2014)

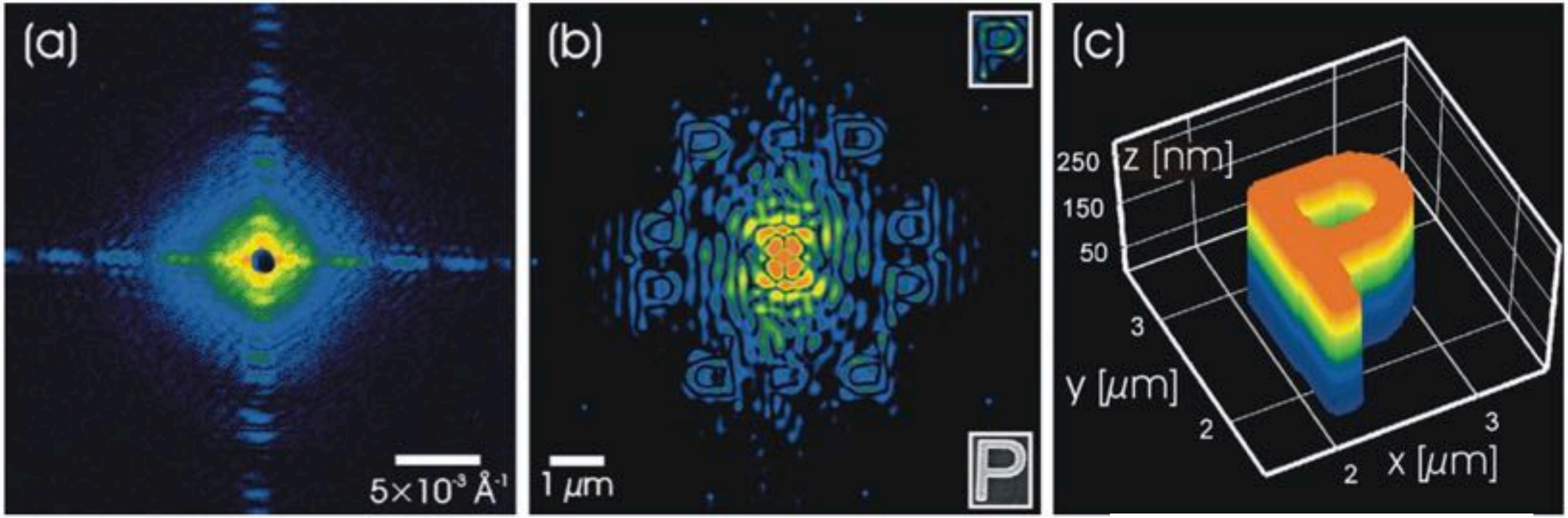
λ = 3.1 nm
α_{max} = 2.86°

Information from a single hologram.
Numerical focussing via propagation.

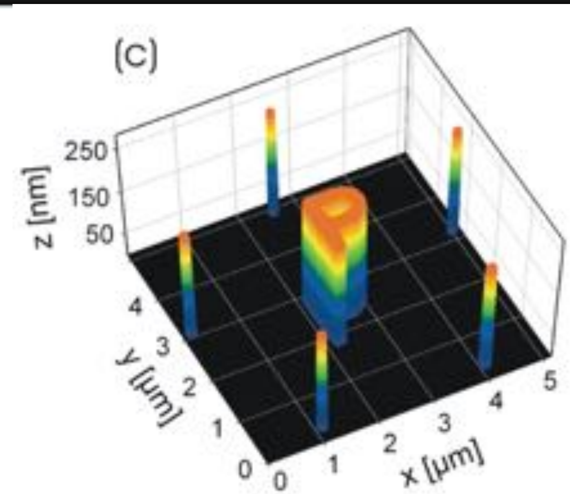
Diffr. limited z resolution = depth of focus = $\frac{D^2_{reference}}{2\lambda}$

FTH with hard x-rays

8 keV, no mask, letter „P“ test structure on open membrane, ≈200 nm gold dots as reference



- amplitude and phase contrast
- also combined with CDI
- 25 nm resolution



L.-M. Stadler et al., PRL **100** 245503 (2008)

These were some of the extra benefits of FTH

- can encode multiple objects in one hologram
- can encode object with different x-ray wavelengths (not shown) or at different times (later)
- scanning the field of view is possible
- 3D information can be encoded
- you obtain the wave field, so you can apply „virtual optics“ in the computer (not shown)

Temporal resolution

single shot images
- light pulse has to be sufficiently short to see the relevant process
- need enough photons during the exposure time

subsequent single shots (= real movie)
- need to record frames fast enough

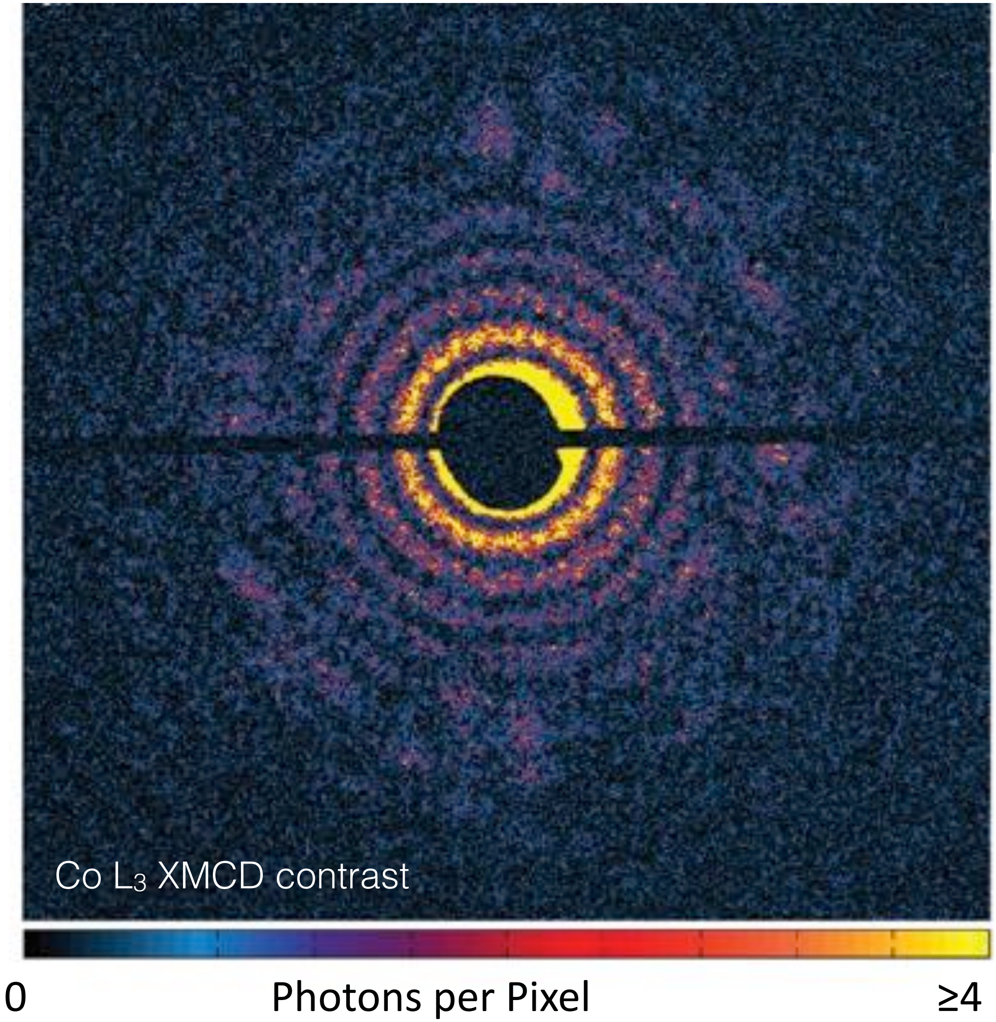
pump-probe
- can look at repetitive processes

https://bda668.wordpress.com

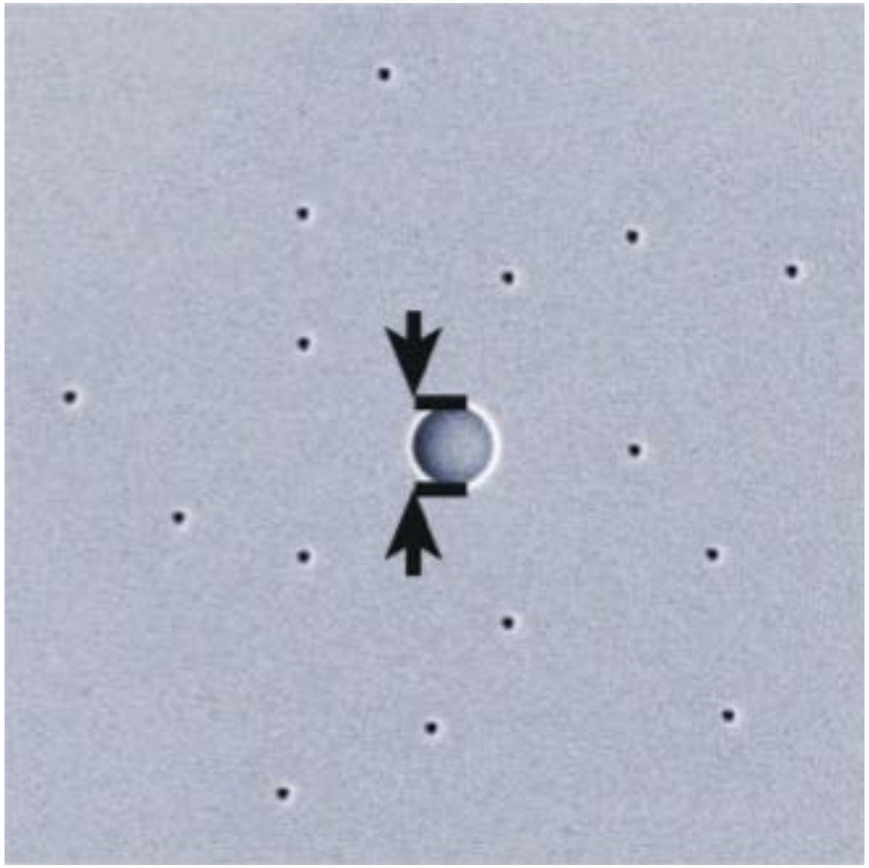
fs single shot FTH image of magnetic domains (SXR @ LCLS)

- multiple references to boost signal
- FTH reconstruction is very robust and does not need many photons, see also W.F. Schlotter APL **89**, 163112 2006

single shot- hologram



reconstruction (magnetization contrast)



single shot at LCLS

- 80 fs pulse
- 778 eV
- 1.5×10^5 photons
- 15 references
- addl. polarizer required as this was prior to delta-Undulator!

T. Wang et al., Phys. Rev. Lett. **108**, 267403 (2012)

Soft vs. hard x-ray Split and Delay technology

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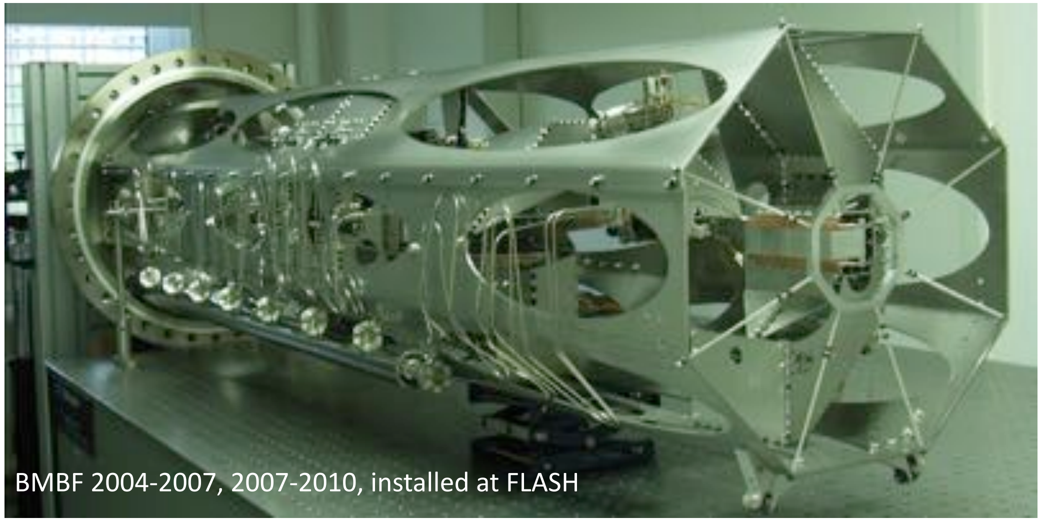
Soft x-rays @ FLASH

(grazing incidence mirrors)



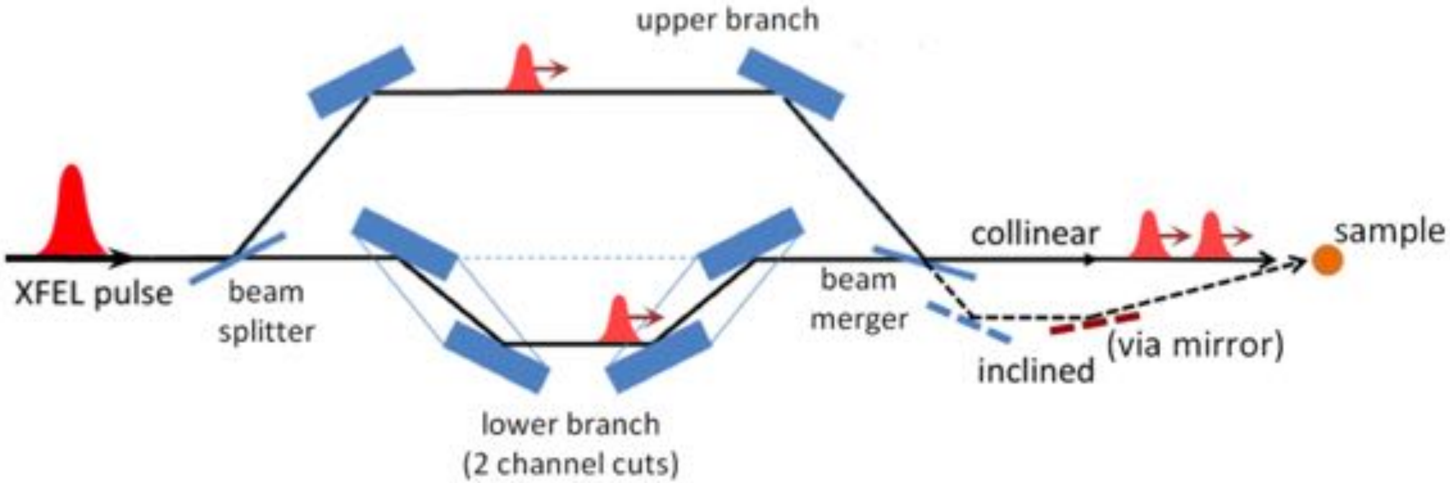
R. Mitzner et al, *Proc. SPIE* **5920**, 59200D (2005) &
Opt. Express 16, 19909–19919 (2008)

Hard X-rays @ XFEL (MID)

(Bragg diffraction, under construction)

E = 5 – 10 keV
Δt = -10 to 800 ps

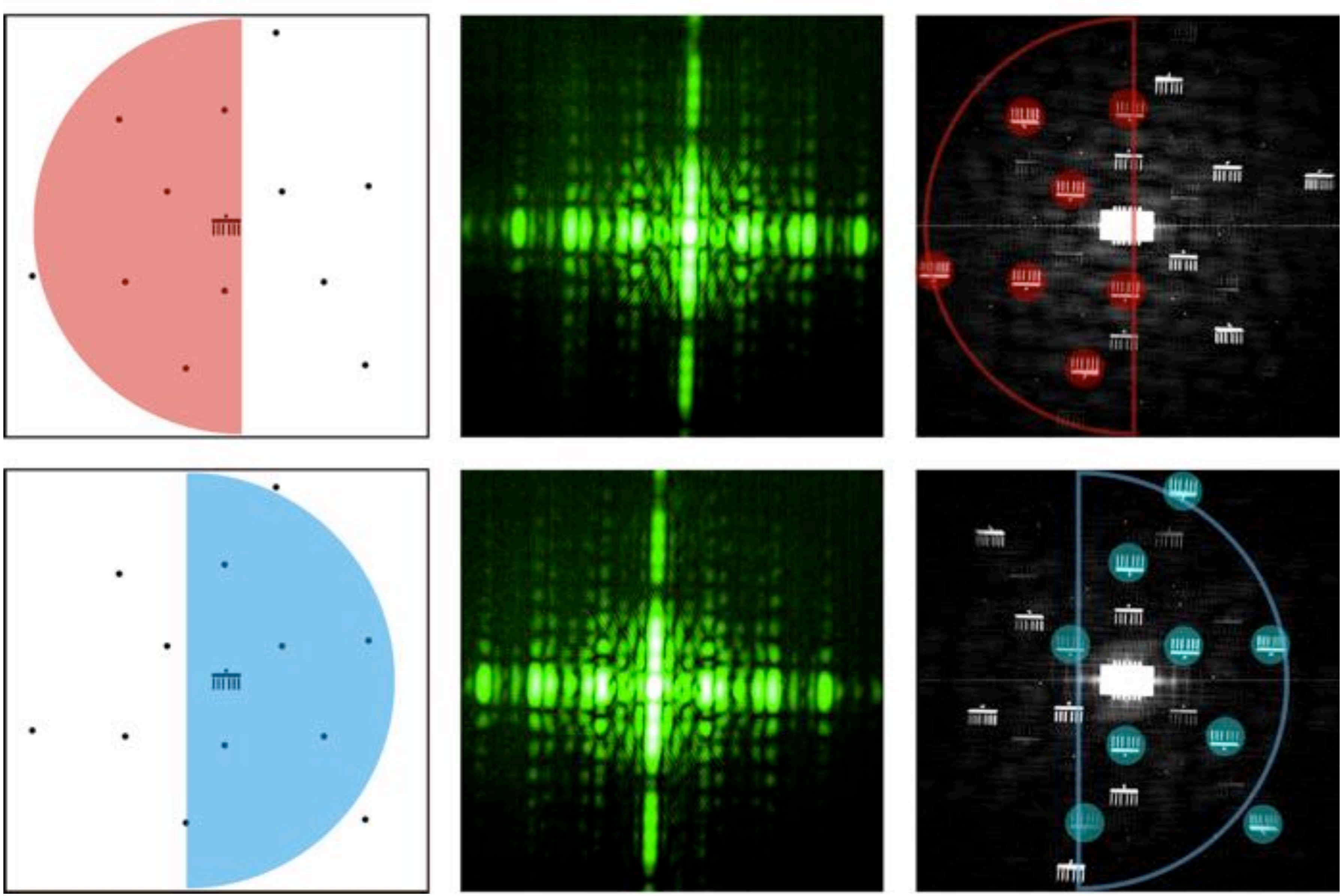
$\Delta t_{XFEL} < 100$ fs



W. Lu et al., AIP Conference Proceedings **1741**, 030010 (2016)

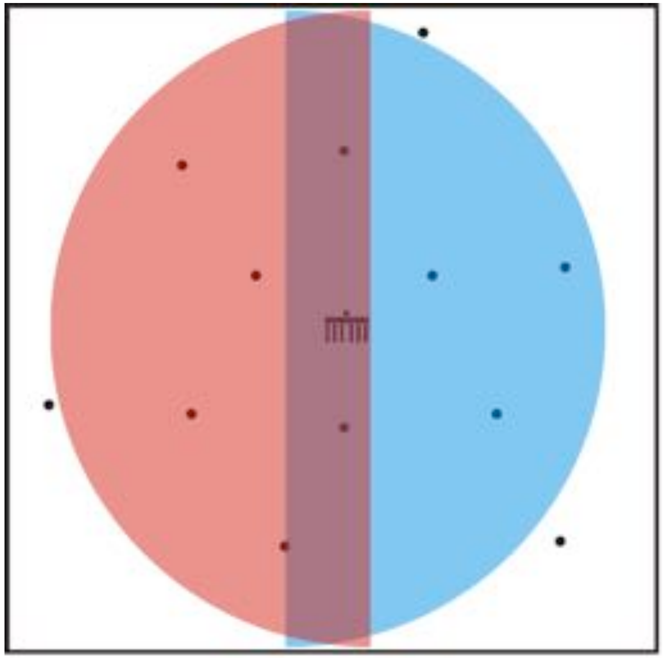
Enable experiments on fs and ps dynamics with high spatial resolution

check: 1 Beam Only

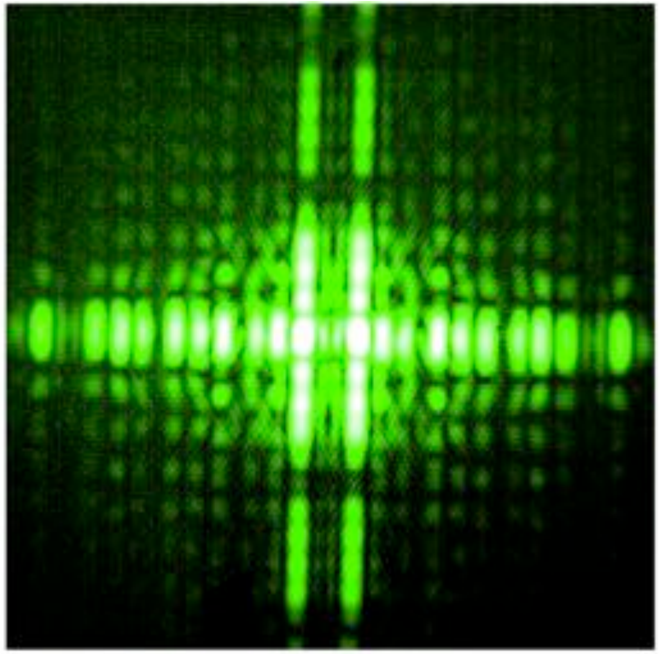


Both beams: map temporal information to space

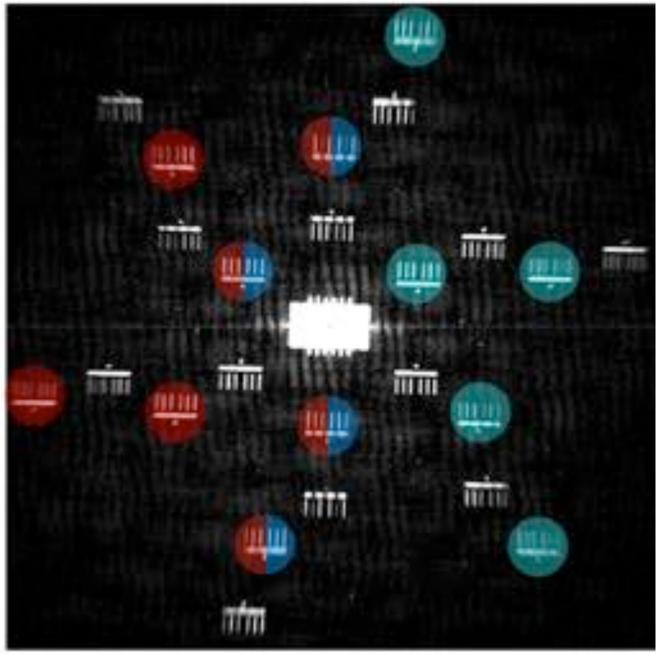
real space object



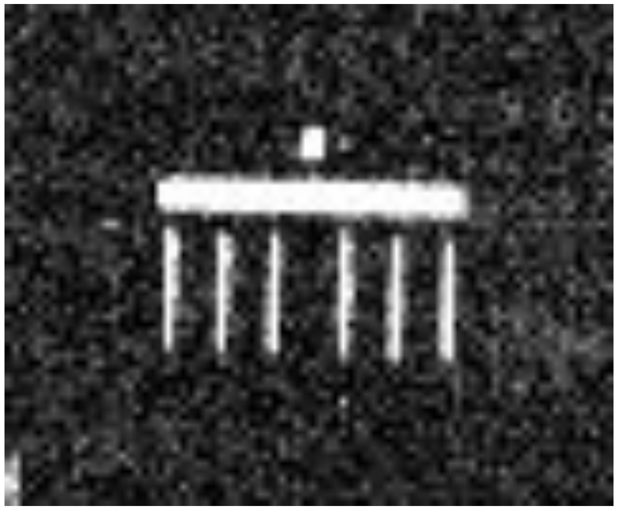
reciprocal space



reconstruction (FFT)

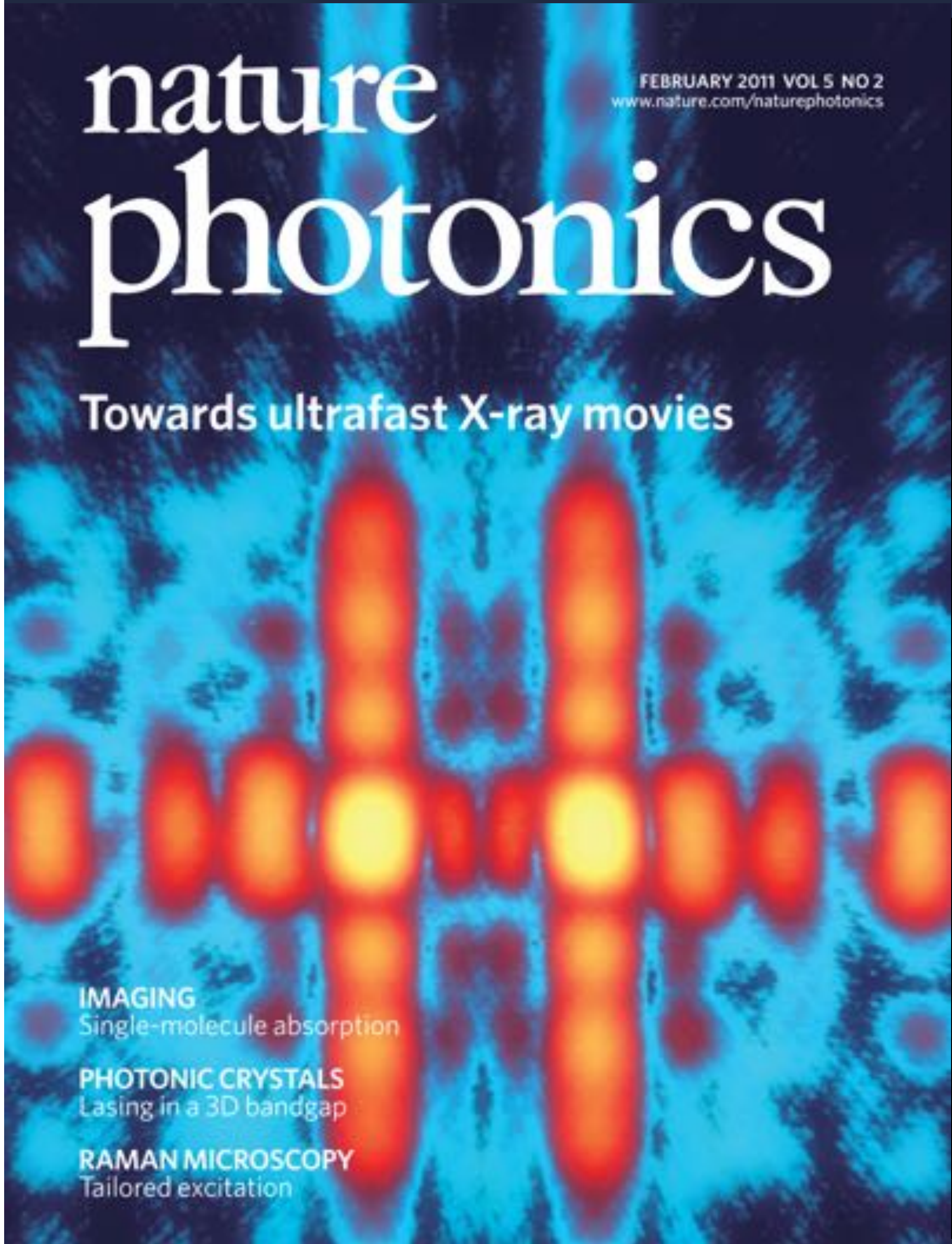


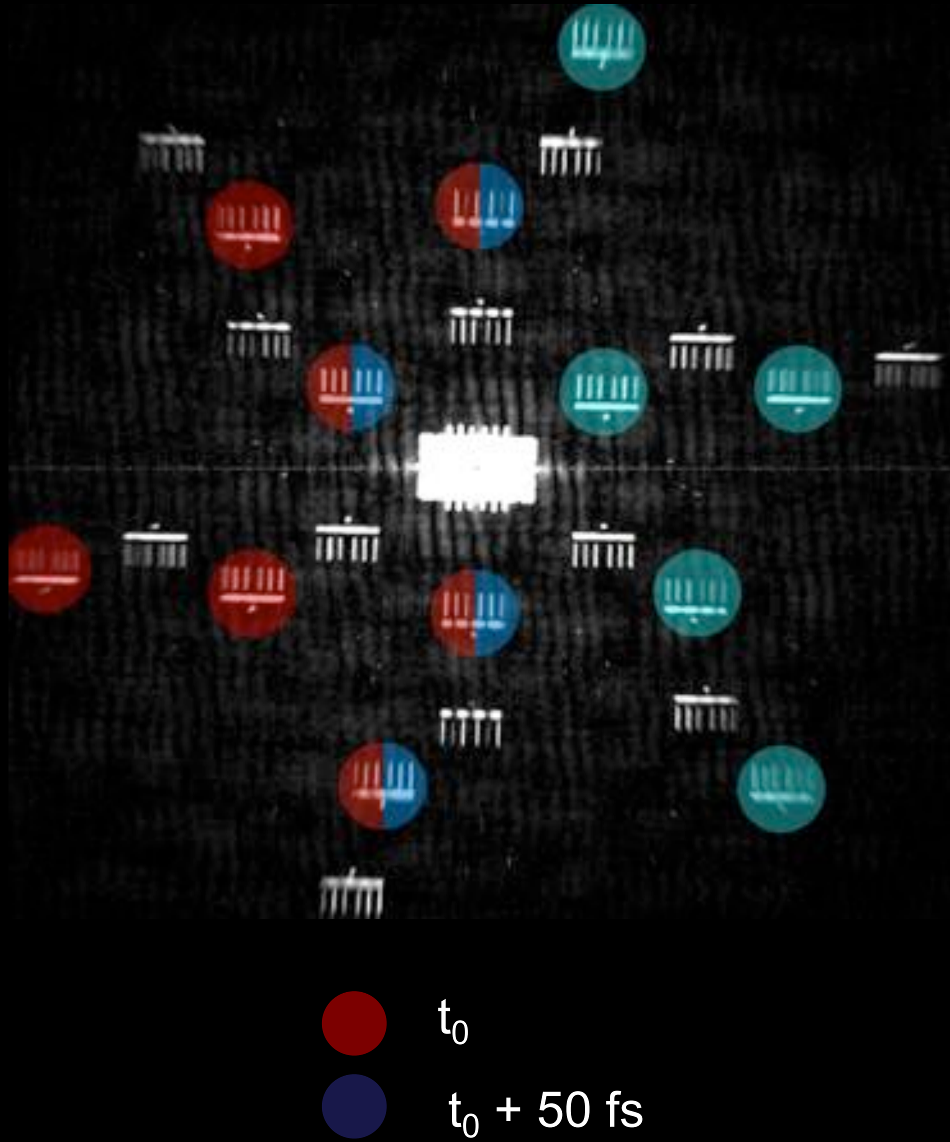
FLASH @ DESY
λ = 23.5 nm



25 fs single shot image

2 Independent, Time-Delayed Images of the Same Object

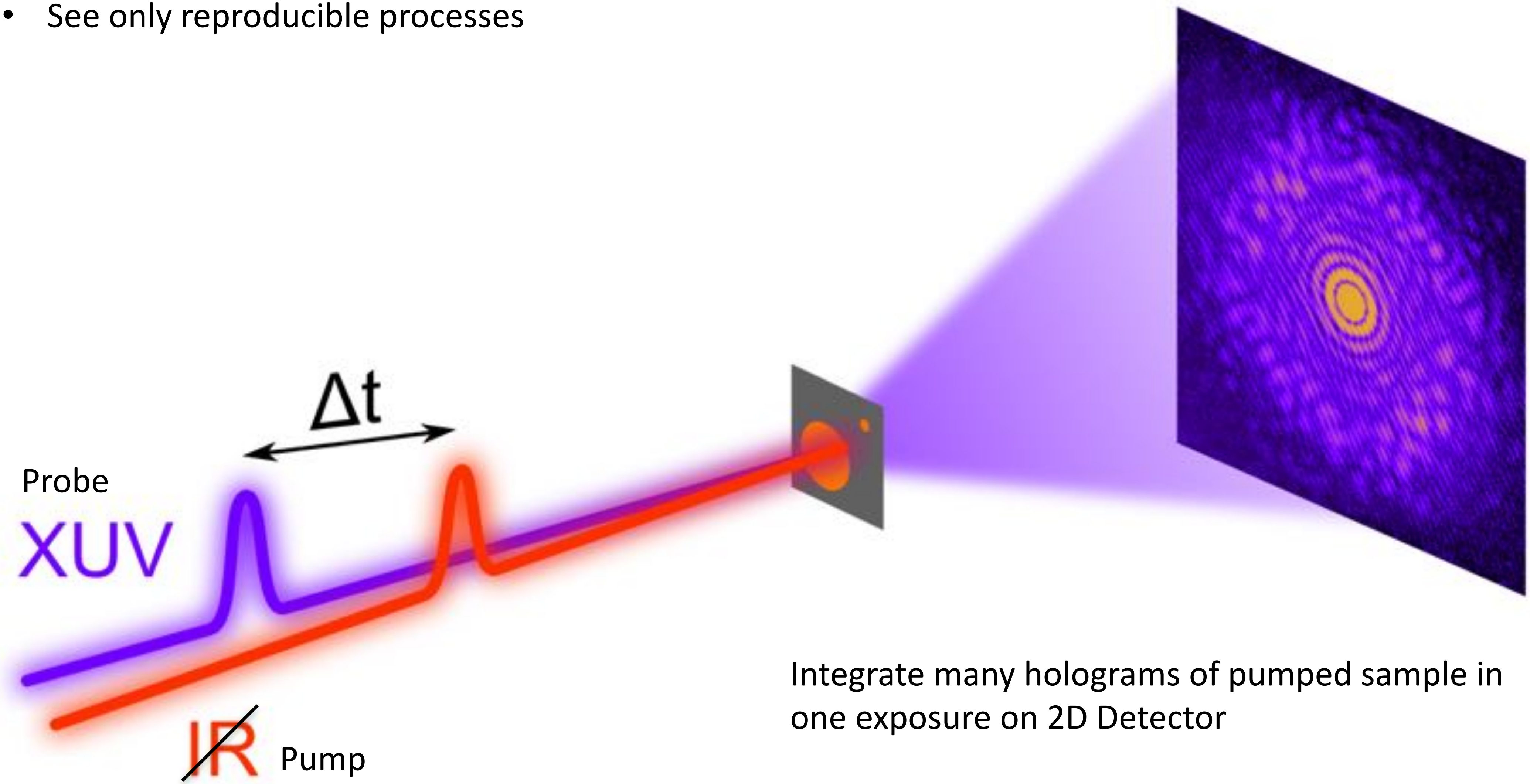




C. Günter et al. Nature Photonics **5**, 99 (2011)

Pump-probe Holography

- Repetitive experiment
- See only reproducible processes



1. experiment: pump = magnetic field

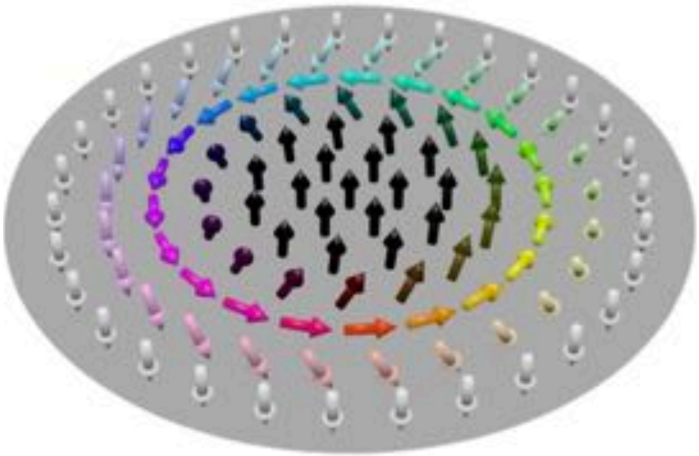
Integrate many holograms of pumped sample in one exposure on 2D Detector

N.B.: single shot images possible at FELs, 1st expt: T. Wang et al, PRL **108** 267403 (2012)

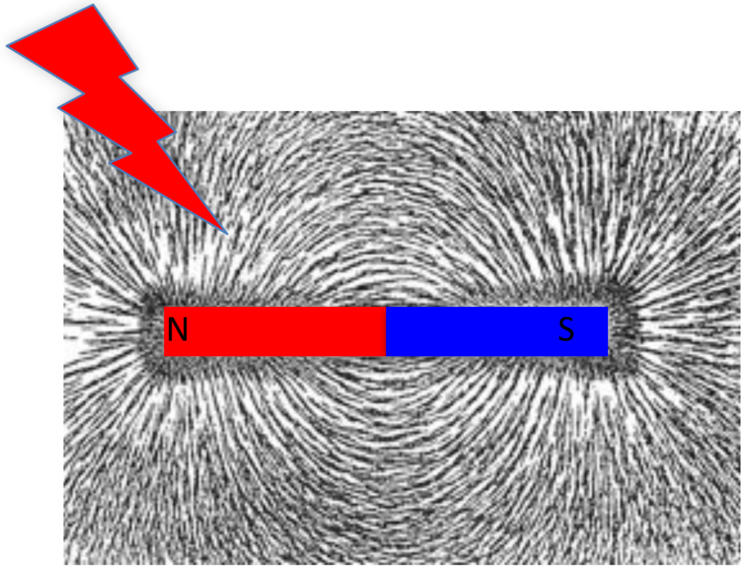
2 Research Examples in Detail

using time-resolved holography (pump-probe)

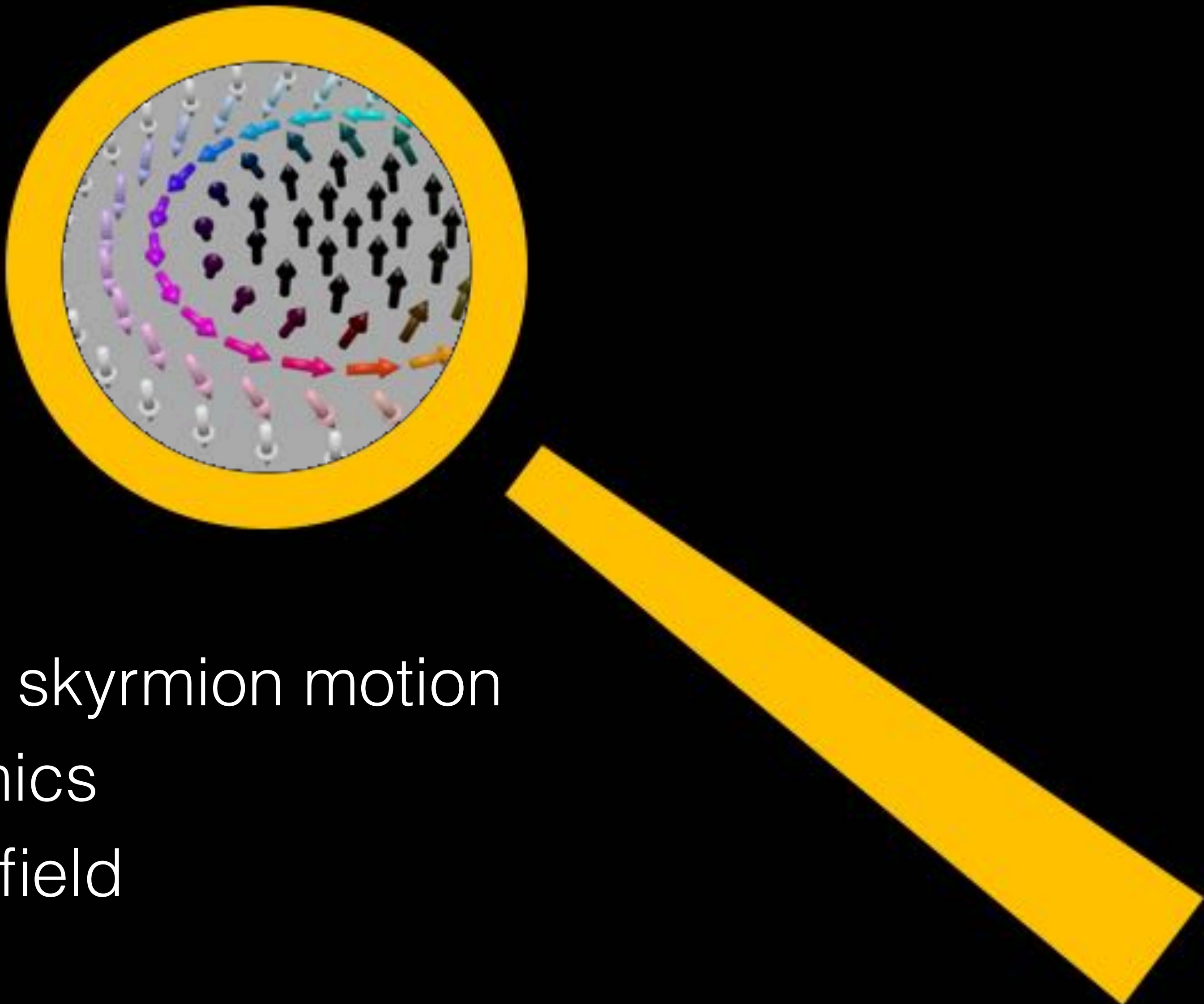
ns, ps
Skyrmion Motion



fs
Electron & Spin Currents

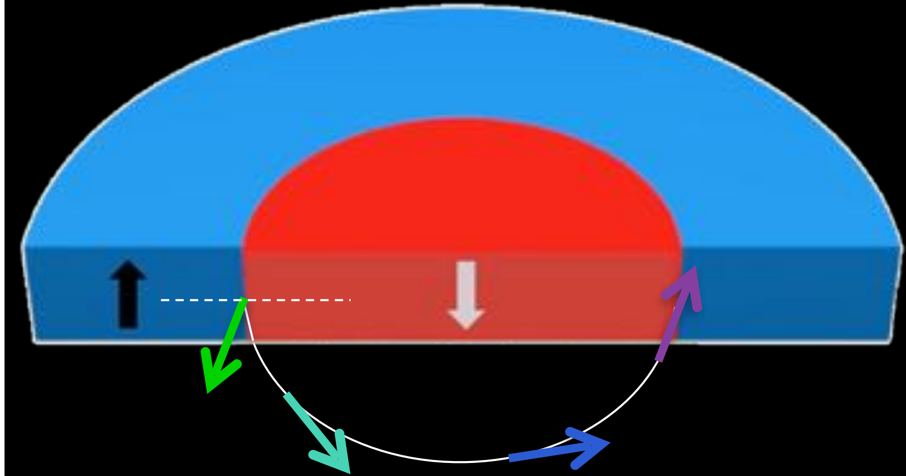


Detailed research example



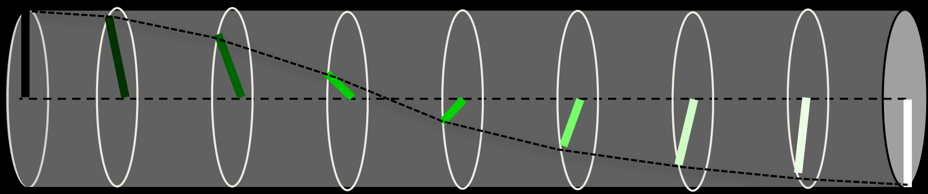
- magnetic skyrmion motion
- ns dynamics
- pump: B-field
- BESSY II

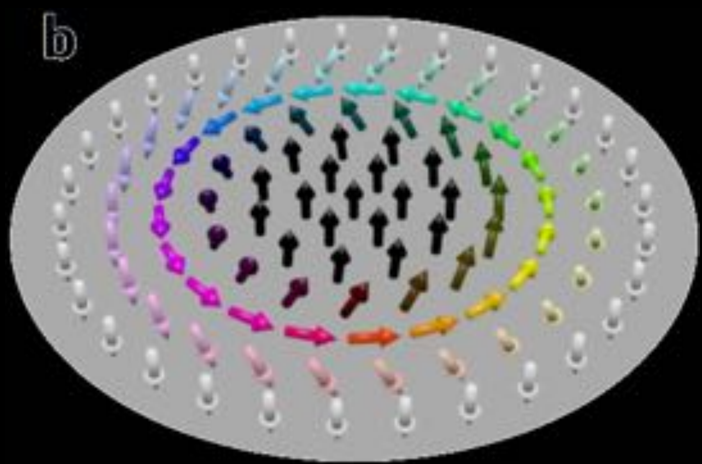
A magnetic bubble is a Skyrmion



Magnetic Bubble
with Bloch wall

in a thin film with perpendicular magnetic anisotropy





Topologically equivalent strcutures ($\mathcal{N} = 1$)

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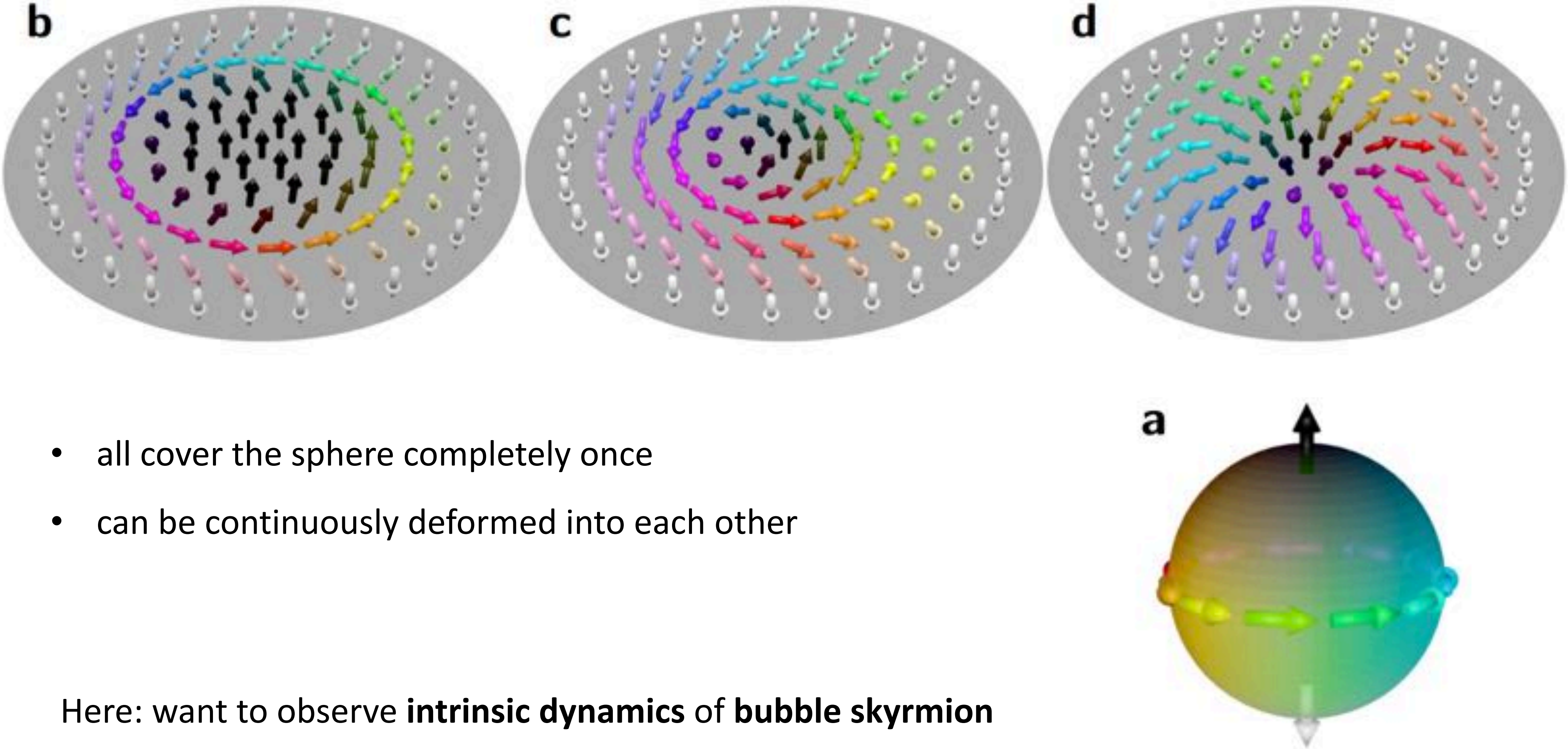
Bubble Skyrmion | **Chiral Skyrmion** | **Hedgehog Skyrmion**

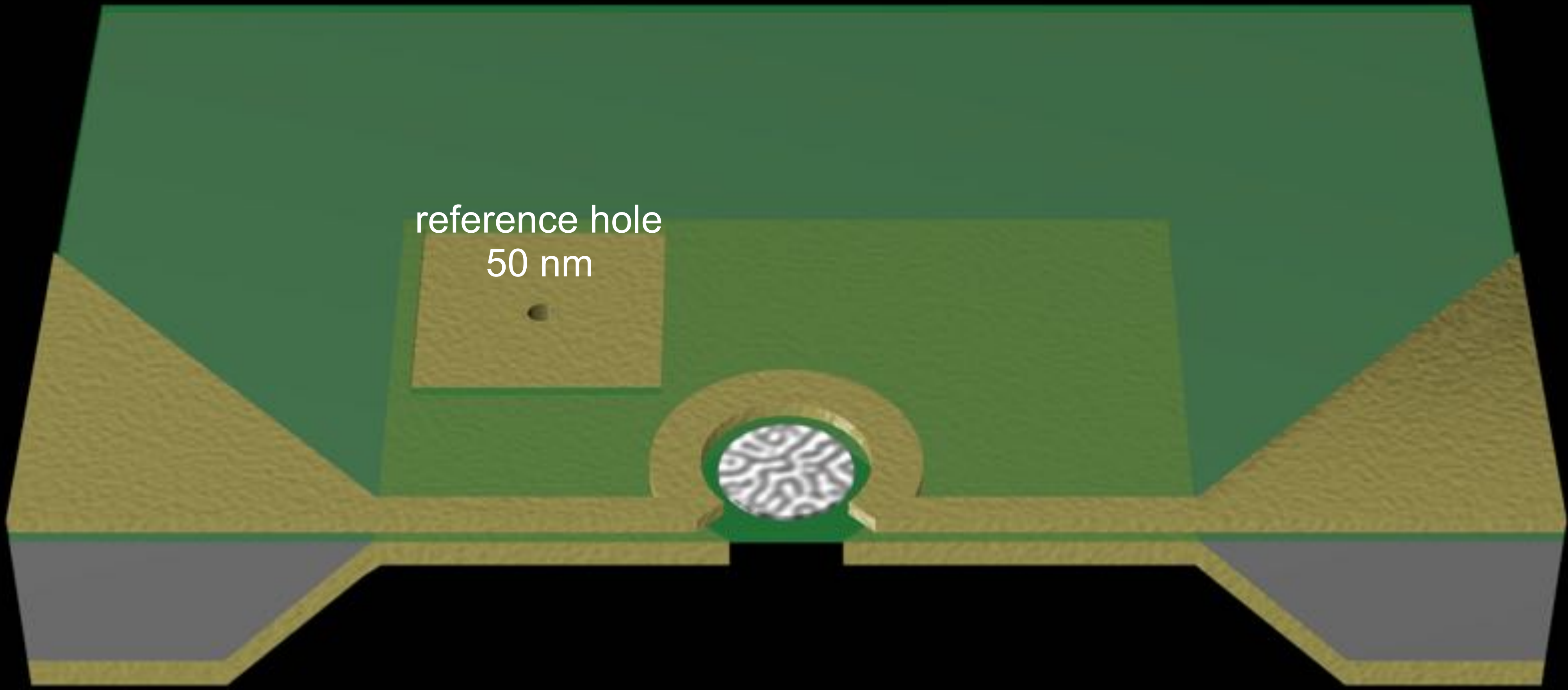


- all cover the sphere completely once
- can be continuously deformed into each other

Here: want to observe **intrinsic dynamics** of **bubble skyrmion**

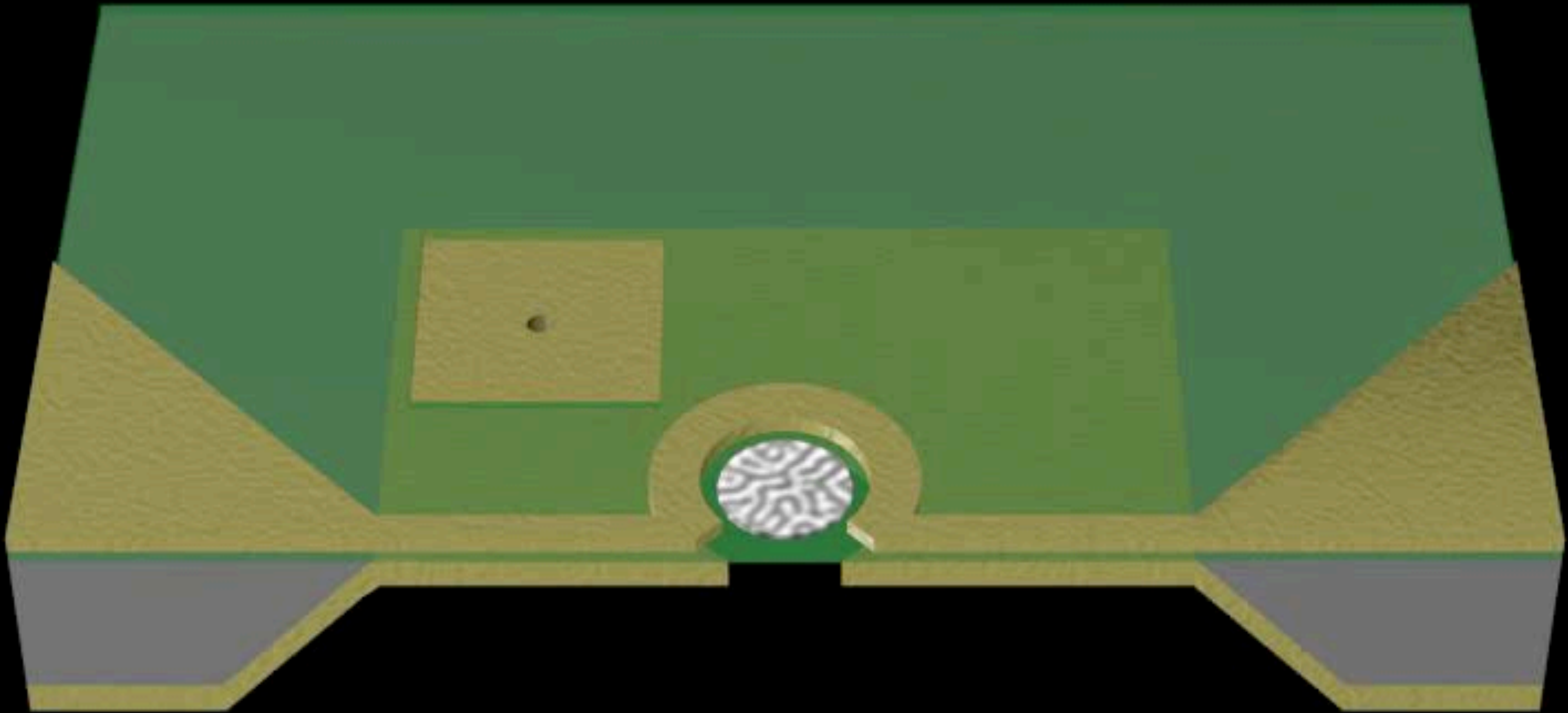
B-pump X-probe FTH: sample design

Investigation of domain wall motion in response to magnetic field pulses



B-pump X-probe FTH: sample design

Investigation of domain wall motion in response to magnetic field pulses



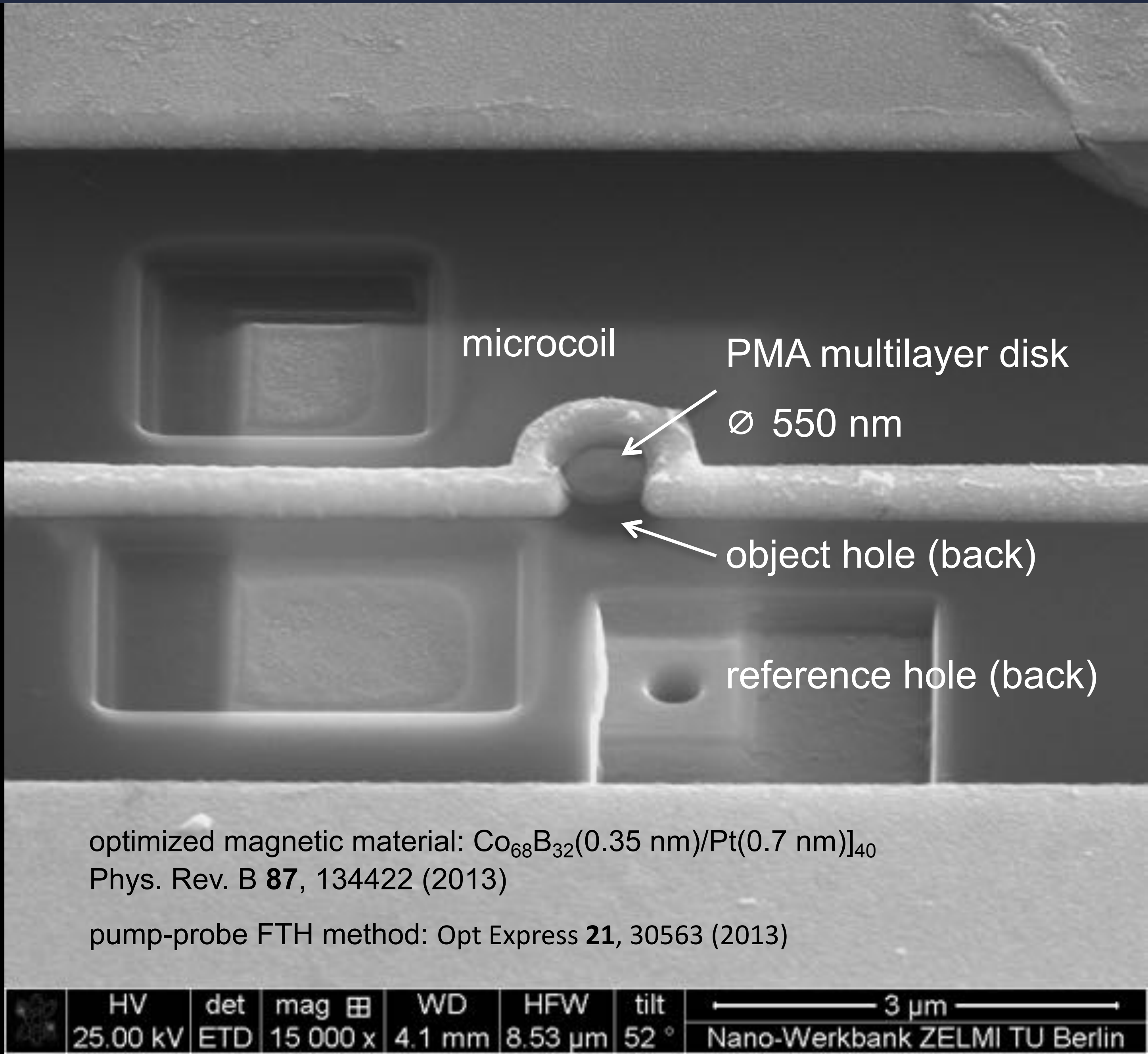
Pushing Magnetic Bubbles

Sample & FTH Geometry

current pulse

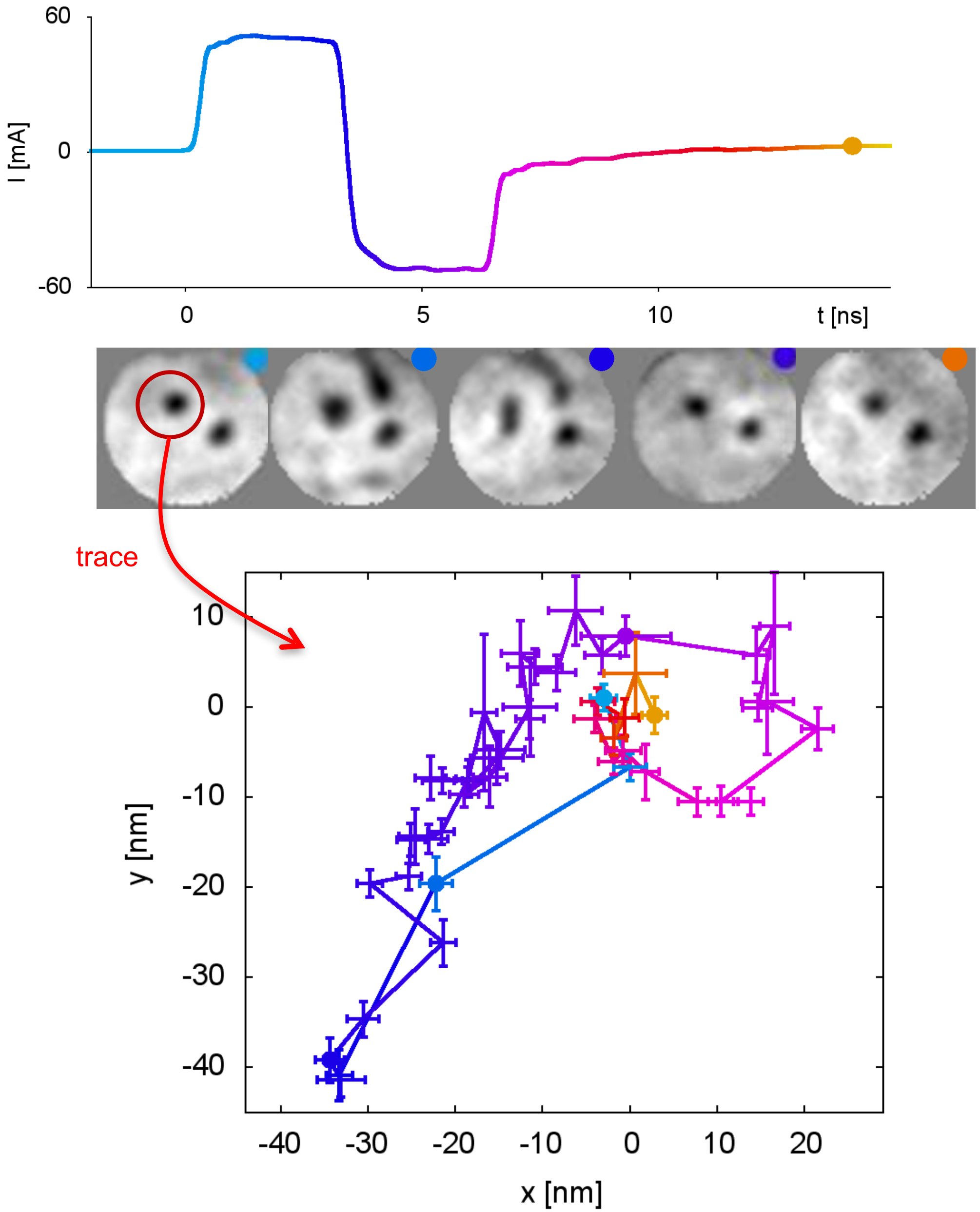
pump-probe
via FTH

BESSY II
single bunch



Observation of gyrotropic motion

- Prepare Skyrmion state in external magnetic field (-125 mT)
- Bipolar field pulse (± 35 mT pulse, 3 ns): generate & annihilate 3rd domain
- Skyrmion is displaced from equilibrium position in response to the stray field
- Relaxation through circular trajectory → gyrotropic motion

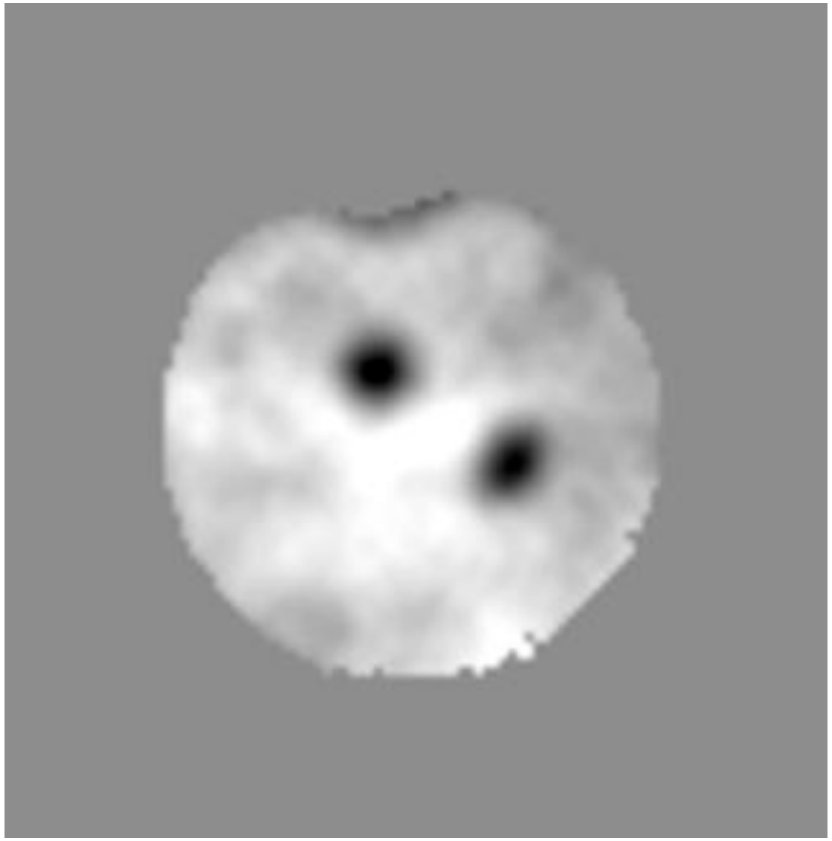


F. Büttner et al., Nature Physics **11**, 225 (2015)

Observation of gyrotropic motion

⌀ 550 nm

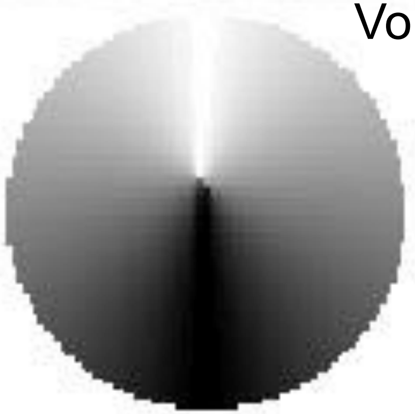
- Prepare Skyrmion state in external magnetic field (-125 mT)
- Bipolar field pulse (± 35 mT pulse, 3 ns): generate & annihilate 3rd domain
- Skyrmion is displaced from equilibrium position in response to the stray field
- Relaxation through circular trajectory → gyrotropic motion



F. Büttner et al., Nature Physics **11**, 225 (2015)

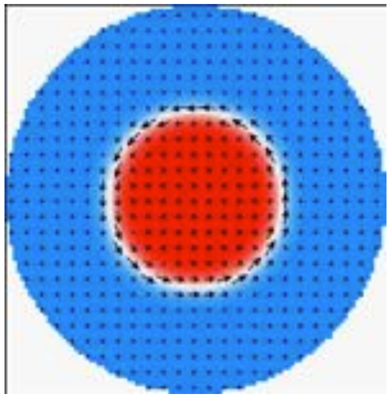
Topology-driven Dynamics

Vortex (N=½)

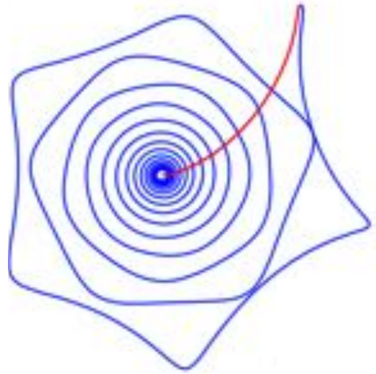


[1]

Skyrmion (N=1) in disk (simulation)



[2]



- Thiele equation of motion for center **R** of *rigid skyrmion:*

$$\mathbf{G} \times \dot{\mathbf{R}} + D\dot{\mathbf{R}} - \partial_{\mathbf{R}} U = 0$$

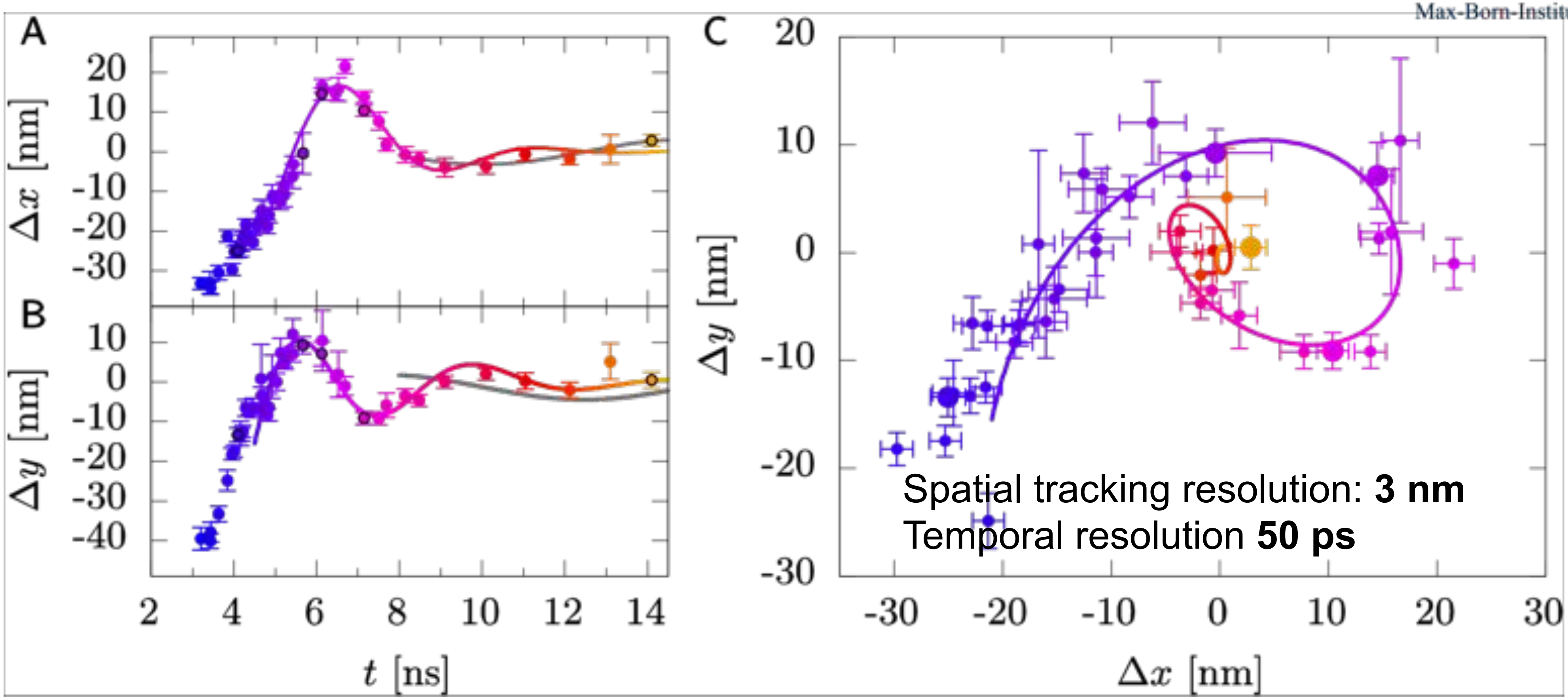
- Gyrovector **G** determined by topological charge N and material properties
- Describes vortex well, fails to describe Skyrmion GHz dynamics
- Skyrmion *deformation* has to be taken into account
- This gives the system an *effective mass:*

$$-M\ddot{\mathbf{R}} + \mathbf{G} \times \dot{\mathbf{R}} + D\dot{\mathbf{R}} - \partial_{\mathbf{R}} U = 0$$

- resulting in two eigenfrequencies [3]

[1] H. Jung et al. Sci. Rep. 1, 59 (2011)
[2] C. Moutafis et al., PRB 79, 224429 (2009)
[3] I. Makhfudz et al., PRL 109, 217201 (2012)

Determination of Skyrmion mass from trajectory



2-frequency fit:

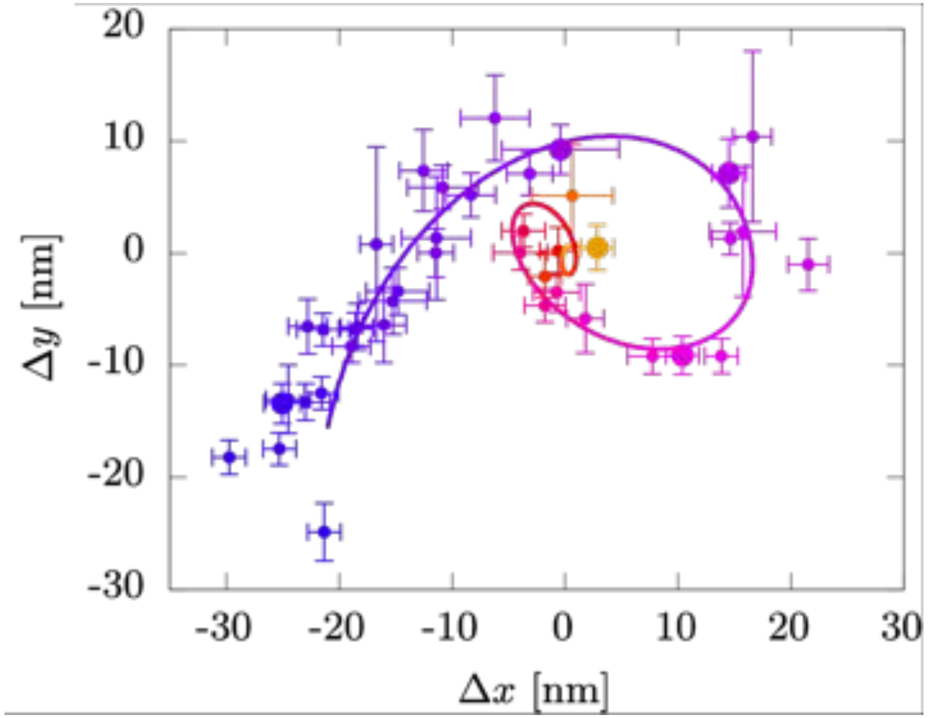
CCW: 1.00 (13) GHz
CW: -1.35 (16) GHz

- existence of eff. mass experimentally confirmed
- large compared to other mag. quasi particles
- topological origin (breathing mode)

F. Büttner et al., Nature Physics **11**, 225 (2015). — work in collaboration with AG Kläui

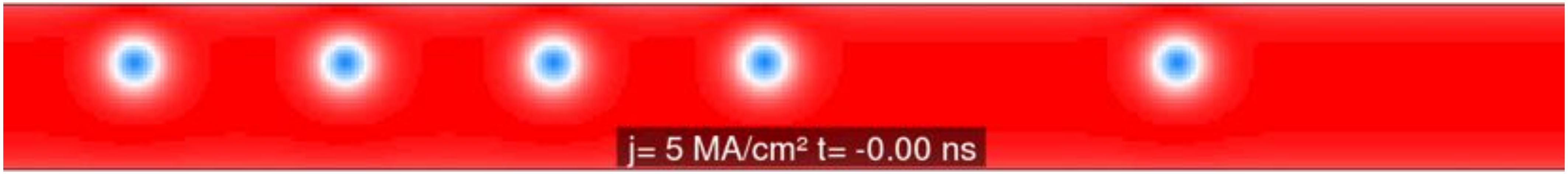
Holographic Imaging Remarks

- **B-pump XMCD-FTH-probe** realized
  @ BESSY II single bunch
- FTH allows for **flexible sample environment** required to prepare the system and trigger its dynamics (here: static field + pulsed field)
- spatial resolution, here: **40 nm**
- **outstanding stability** of FTH allows very precise tracking of absolute position (≈**3 nm**), neither vibrations nor drift are are a problem
- **sensitivity**, here: Magnetization from 9.5 nm Co in 400 nm material observed in transmission. Recent Experiments at PETRA III: **0.5 nm**.



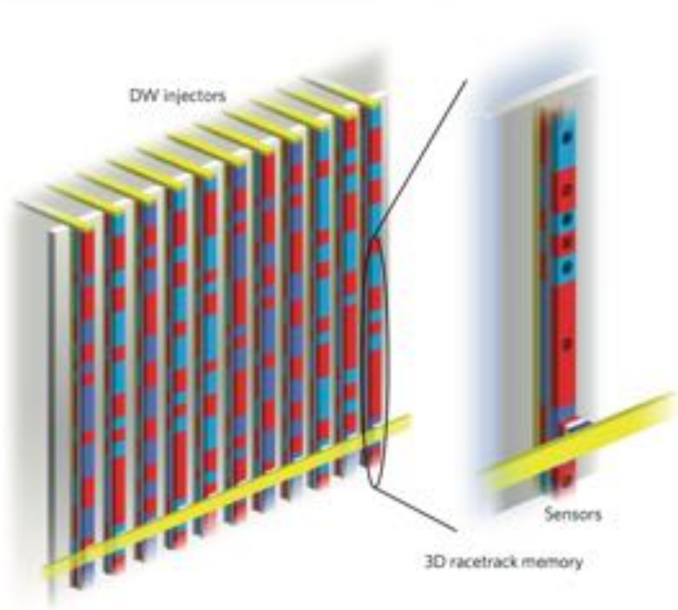
Skyrmion Racetrack Memory

- Skyrmions can be moved in wires, e.g. via Spin Transfer Torque (here: Simulation)



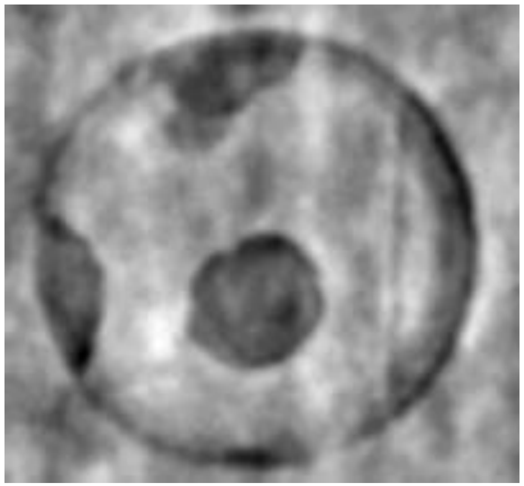
A. Fert et al. Nature Nanotechnology **8**, 152–156 (2013)

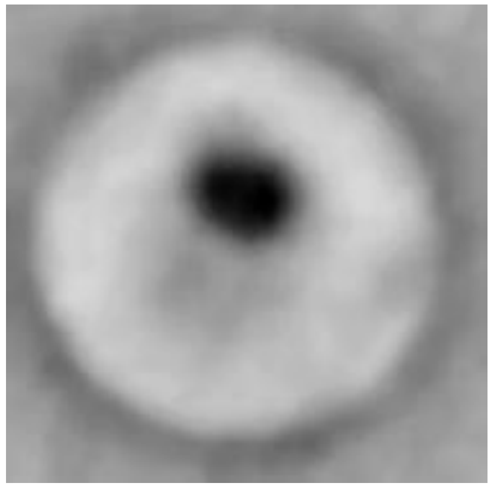
- Racetrack memory concept



S. Parkin & S.-H. Yang Nat. Nanotech. **10**, 195–198 (2015)

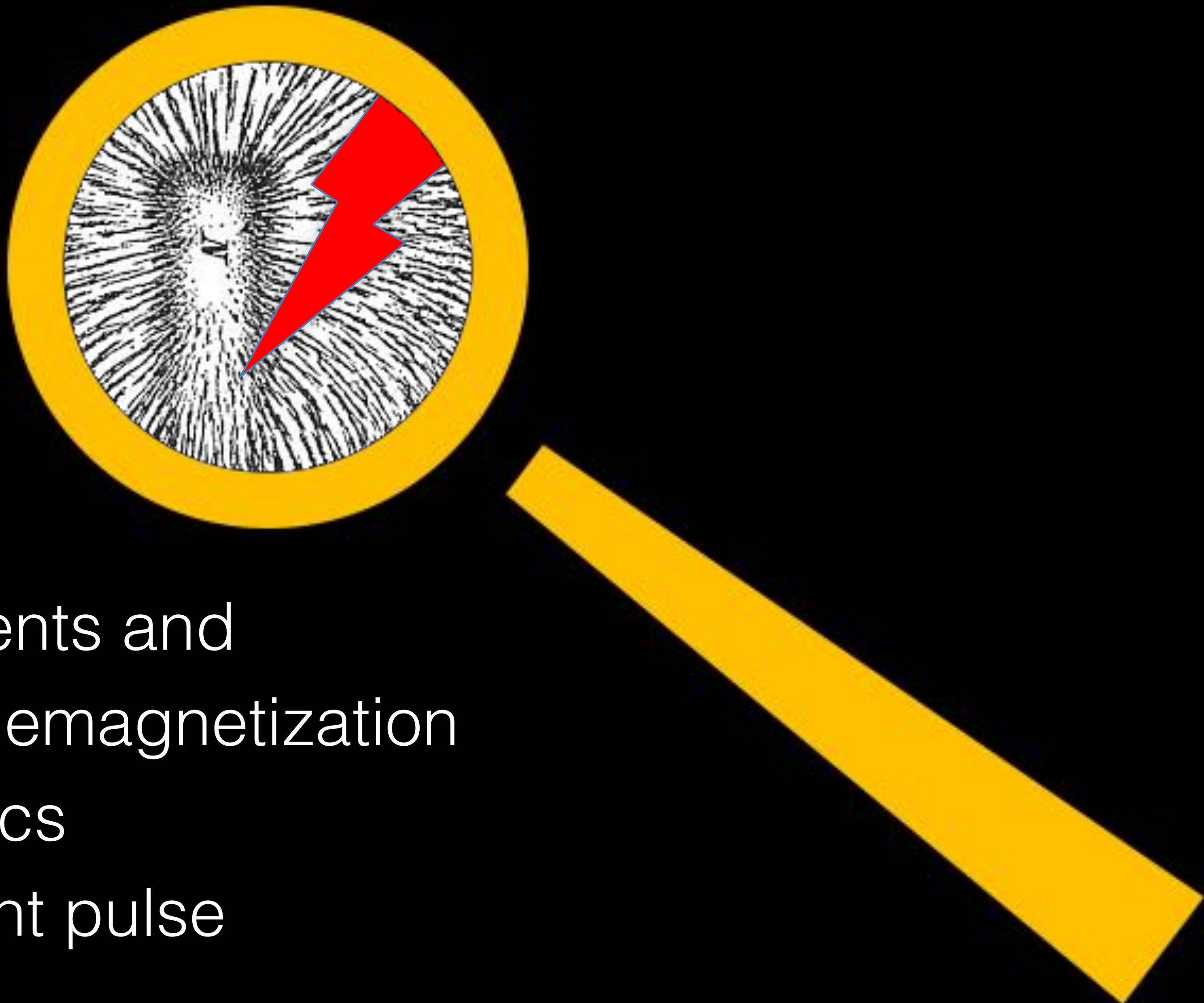
- **Our current work:**
  Controlled generation of Skyrmions via Spin-Hall-Effect





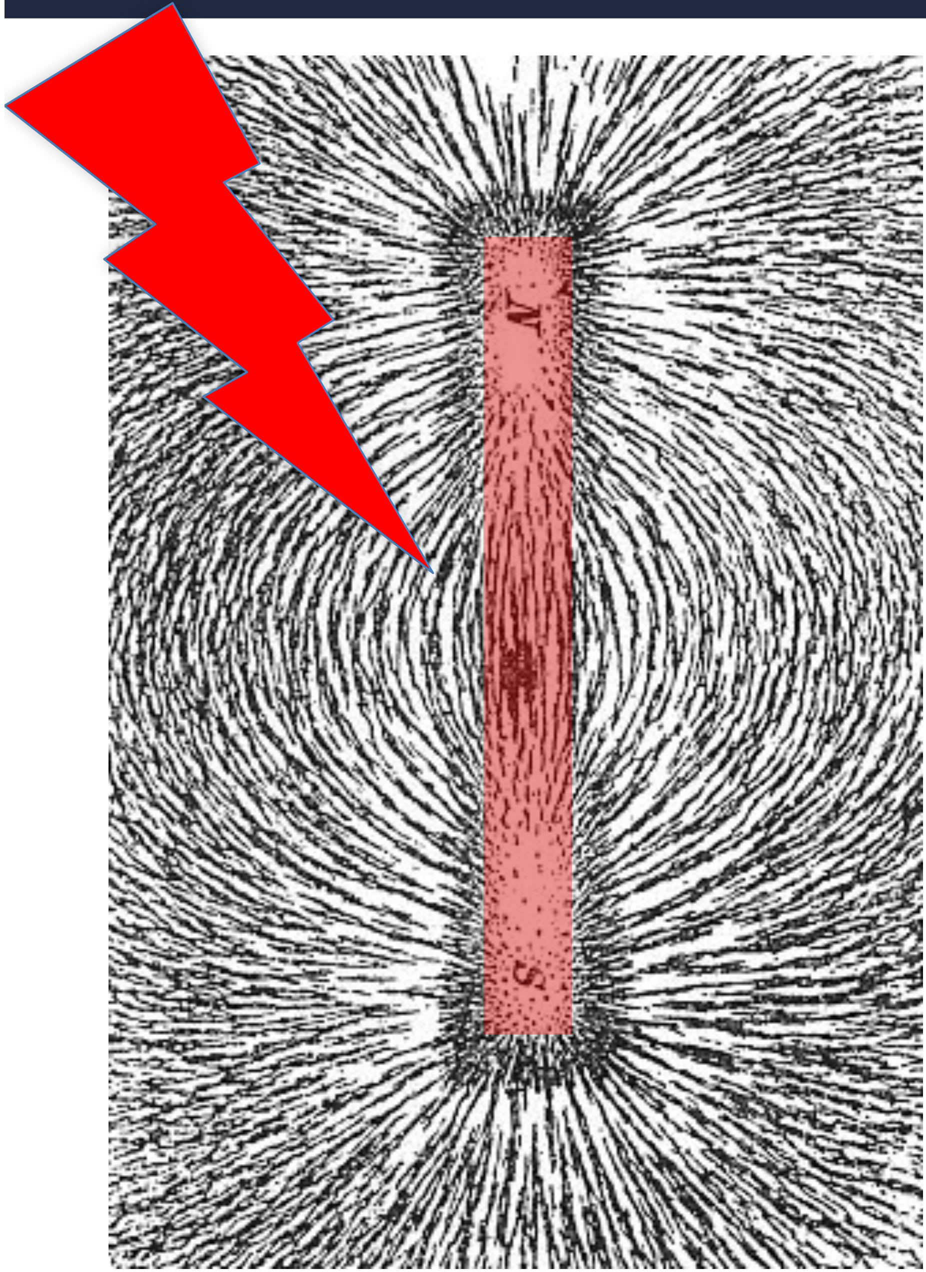
F. Büttner et al. (arXiv:1705.01927) FOV ⌀ 1 µm

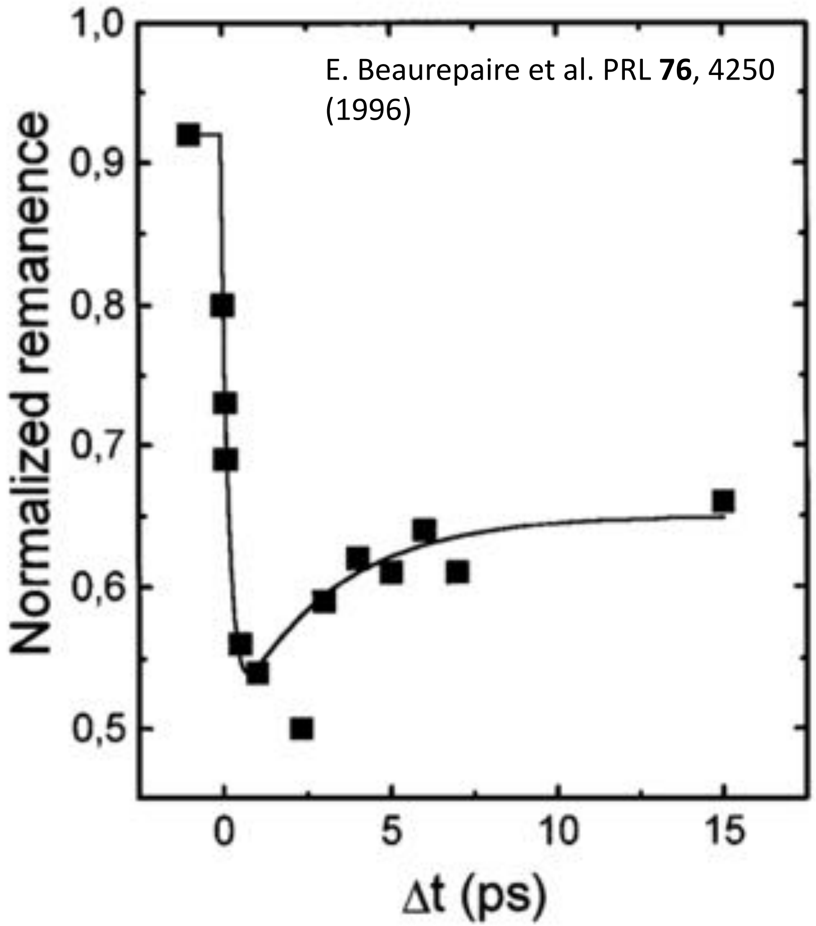
Detailed research example



- Spin currents and ultrafast demagnetization
- fs dynamics
- pump: light pulse
- FERMI

fs Dynamics: Ultrafast Optical Demagnetization





Magnetization can be „destroyed“ on an ultrafast time scale

Relevant Elementary Processes ?

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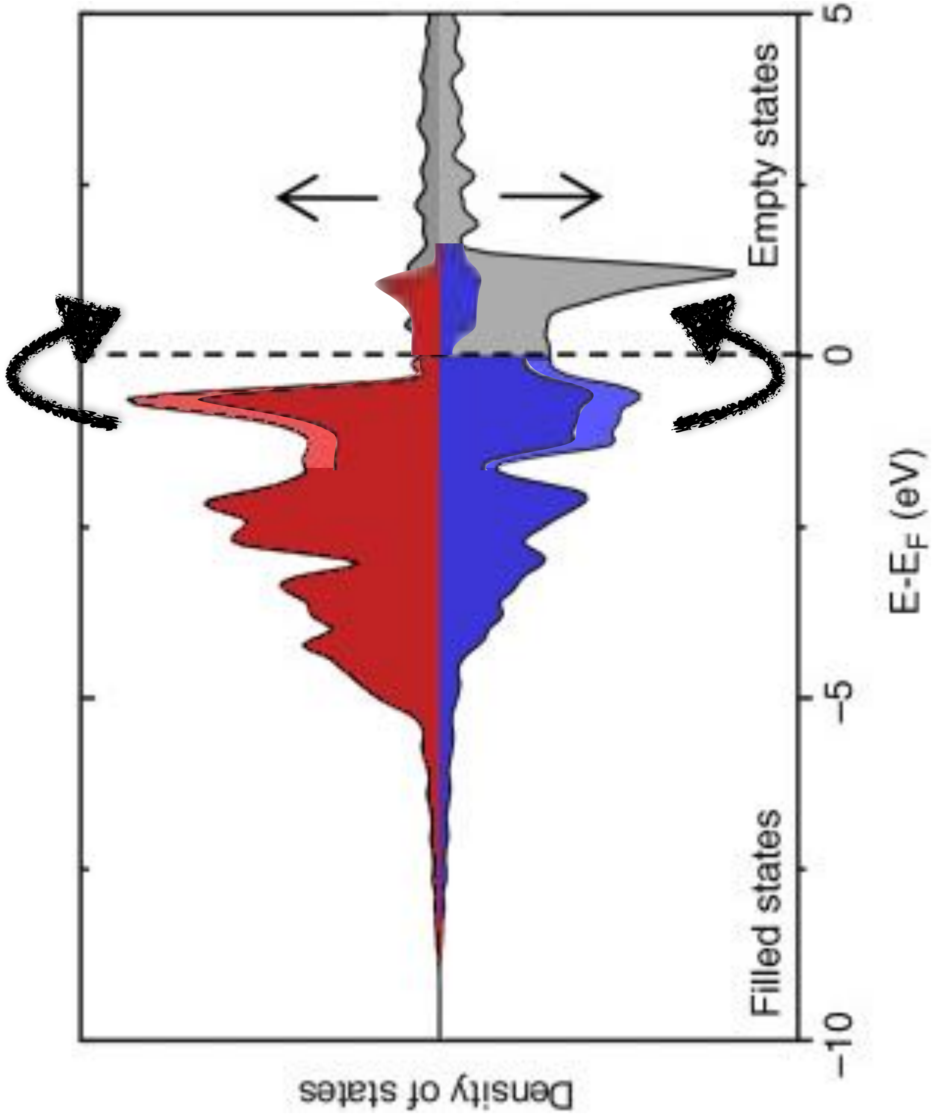
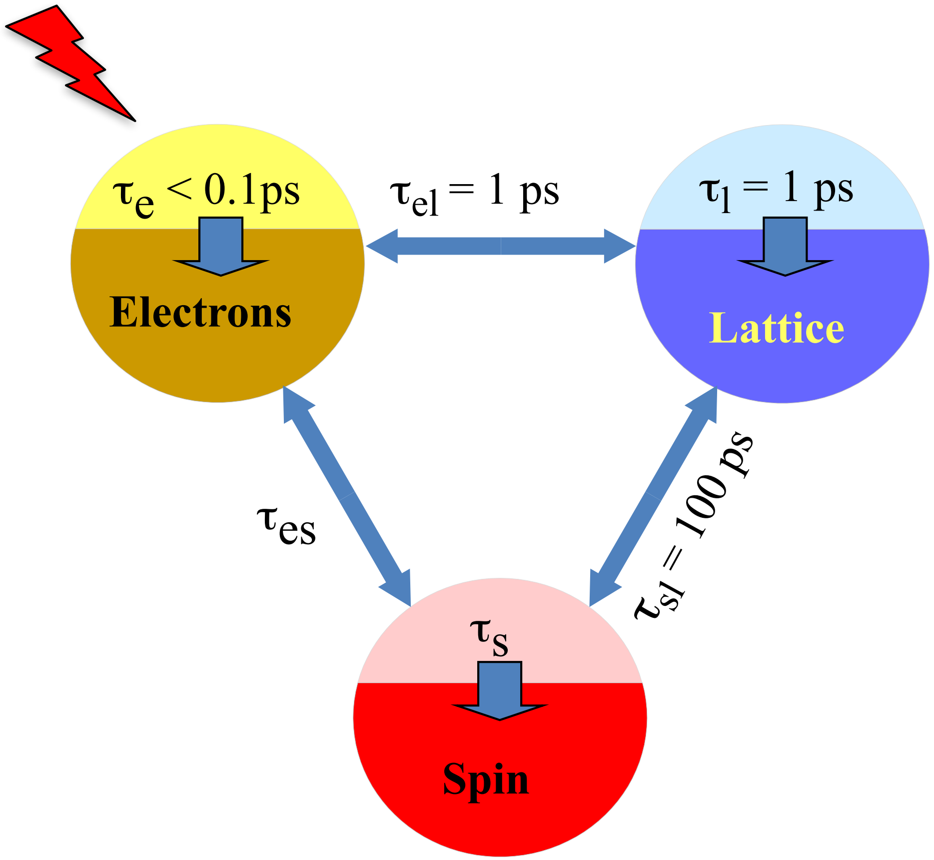
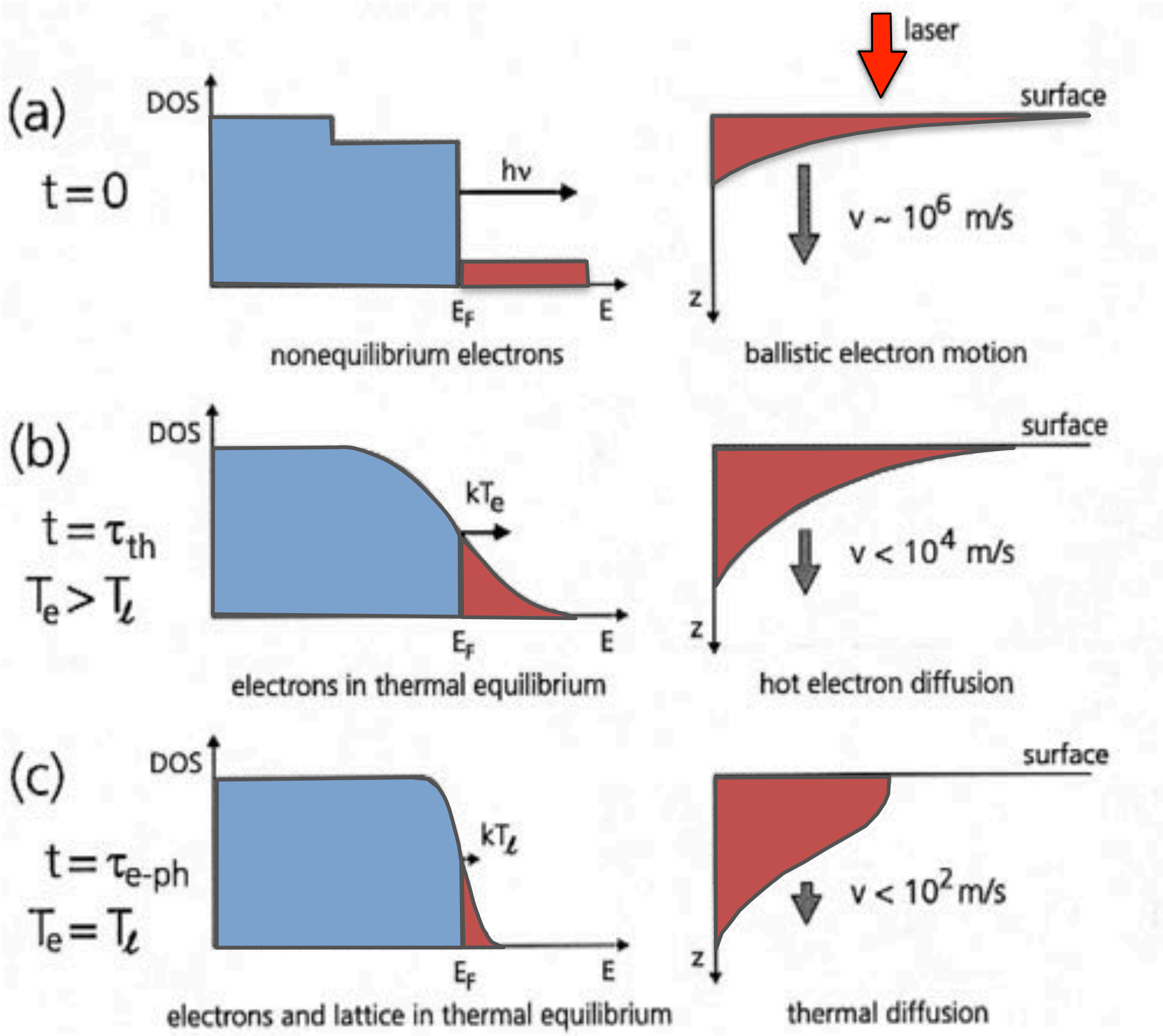




relevance of various mechanisms is an active area of research

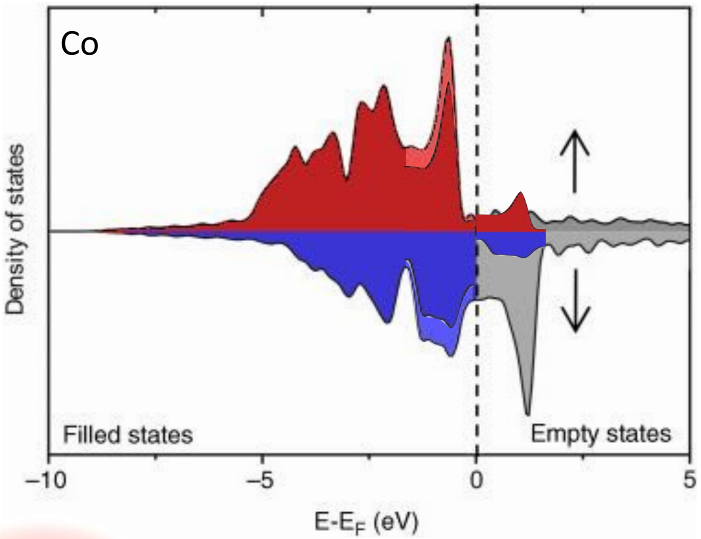
(Hot) Electron Transport

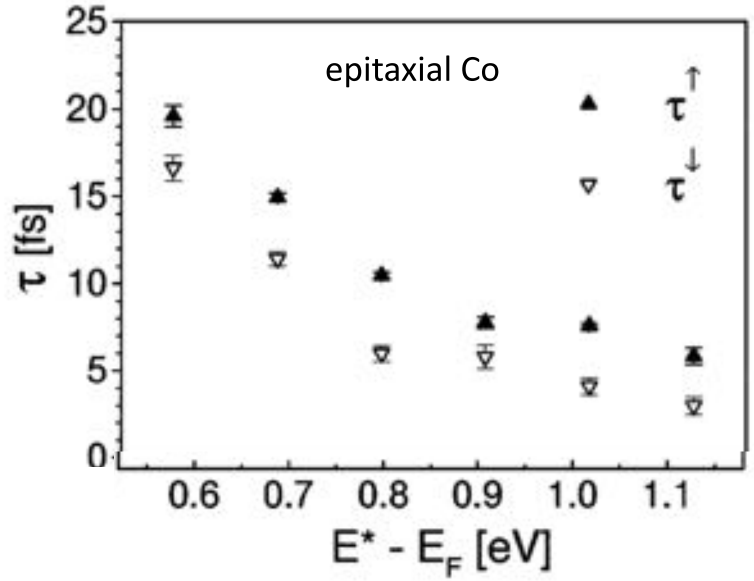


J. Hohlfeld et al., Chem. Phys. **251**, 237 (2000)

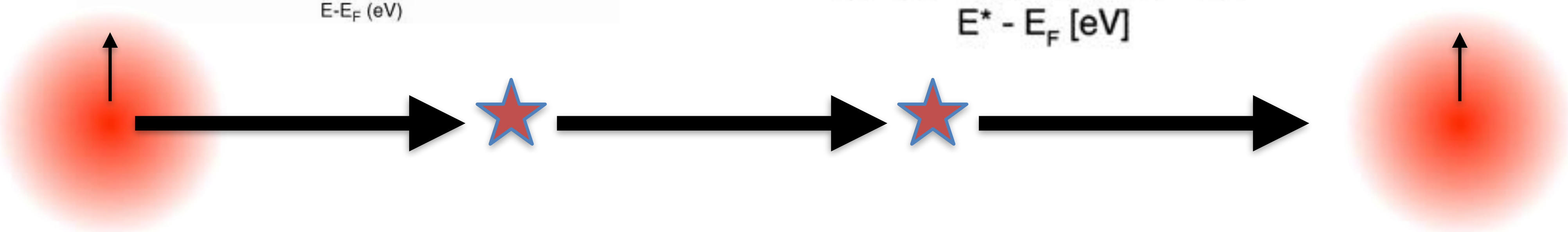
Spin-dependent transport of hot electrons

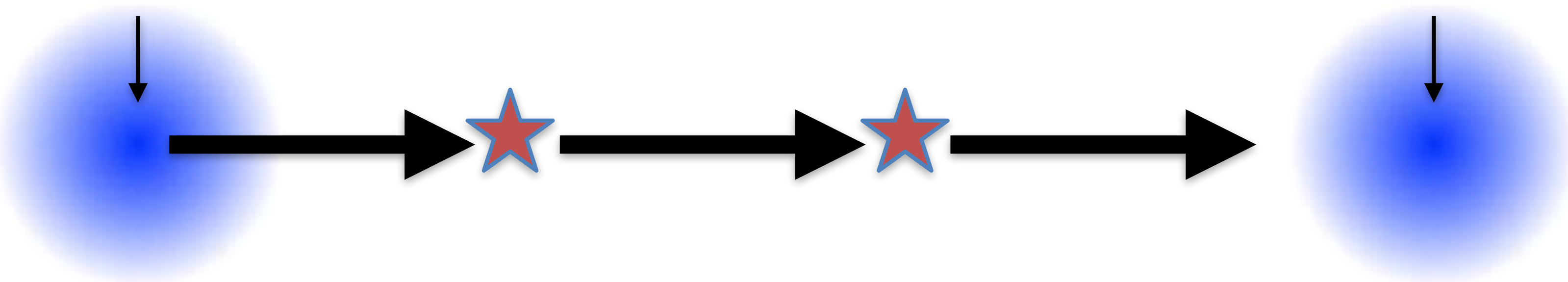
inelastic mean free path different for ↑ and ↓





M. Aeschlimann et al, PRL **79**, 5158 (1997)





↗ Mott 2-current model

+ Interfaces:

↗ Spin Valves

↗ GMR Sensor

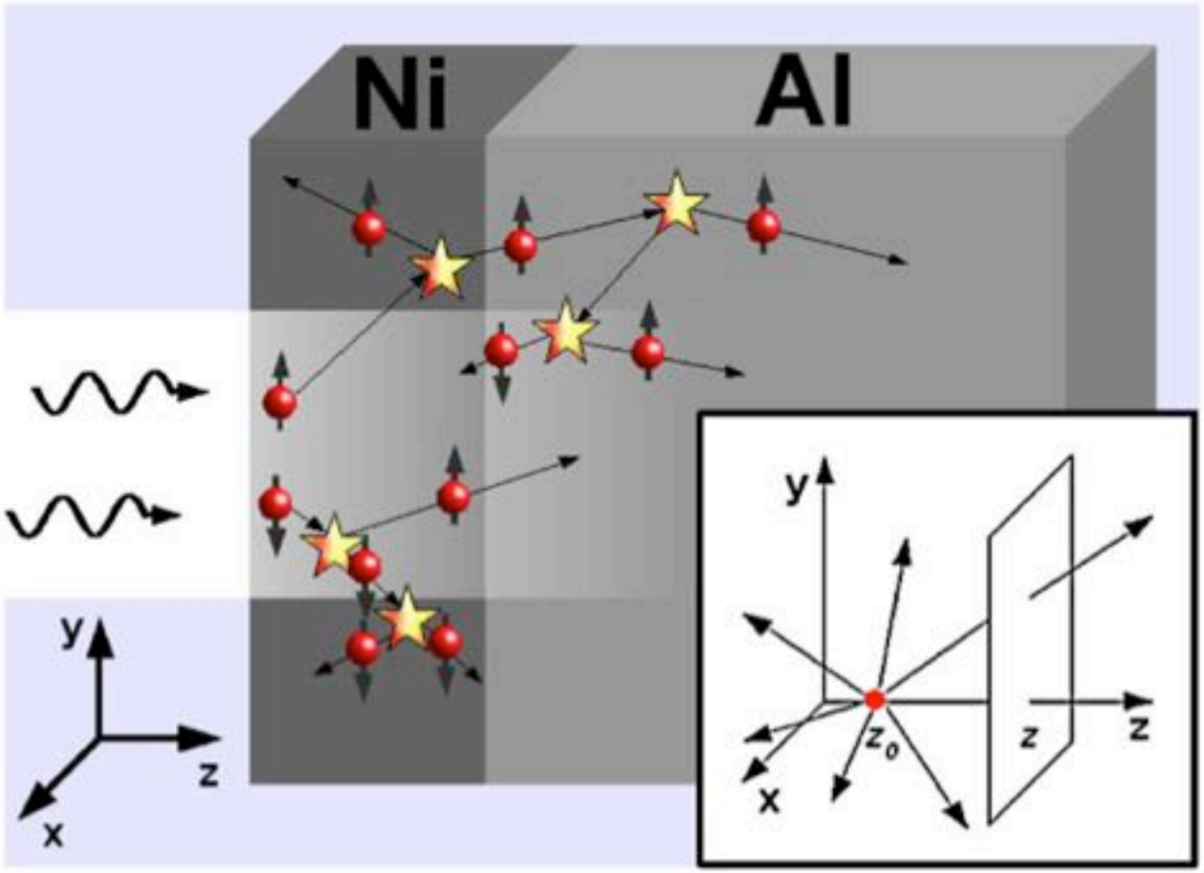
Hot electrons can transport magnetization

due to *spin-dependent mean free path* for majority vs. minority electrons

PRL **105**, 027203 (2010) PHYSICAL REVIEW LETTERS week ending 9 JULY 2010

Superdiffusive Spin Transport as a Mechanism of Ultrafast Demagnetization

M. Battiato,* K. Carva,† and P. M. Oppeneer



initially ballistic,
later diffusive

Lateral transport: no chemical interface

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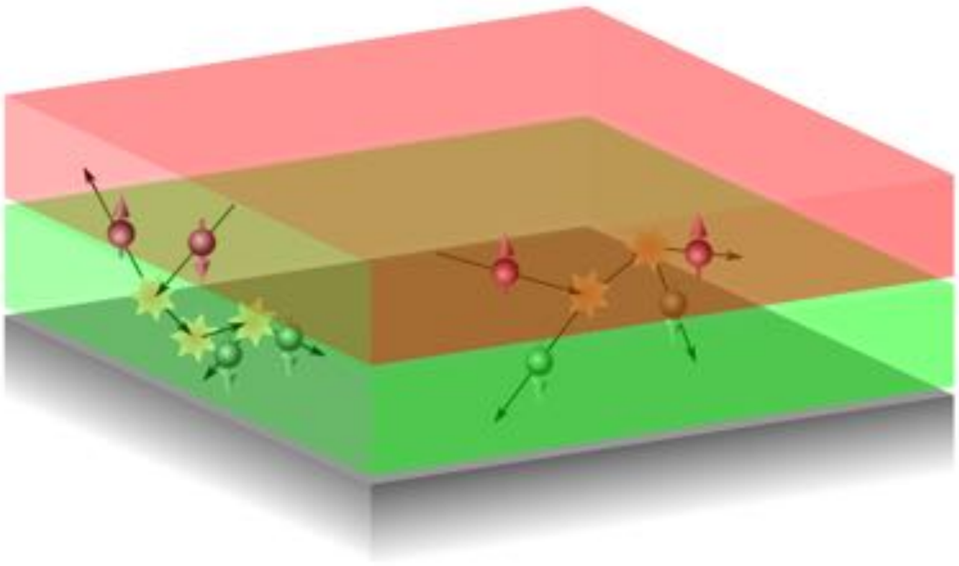
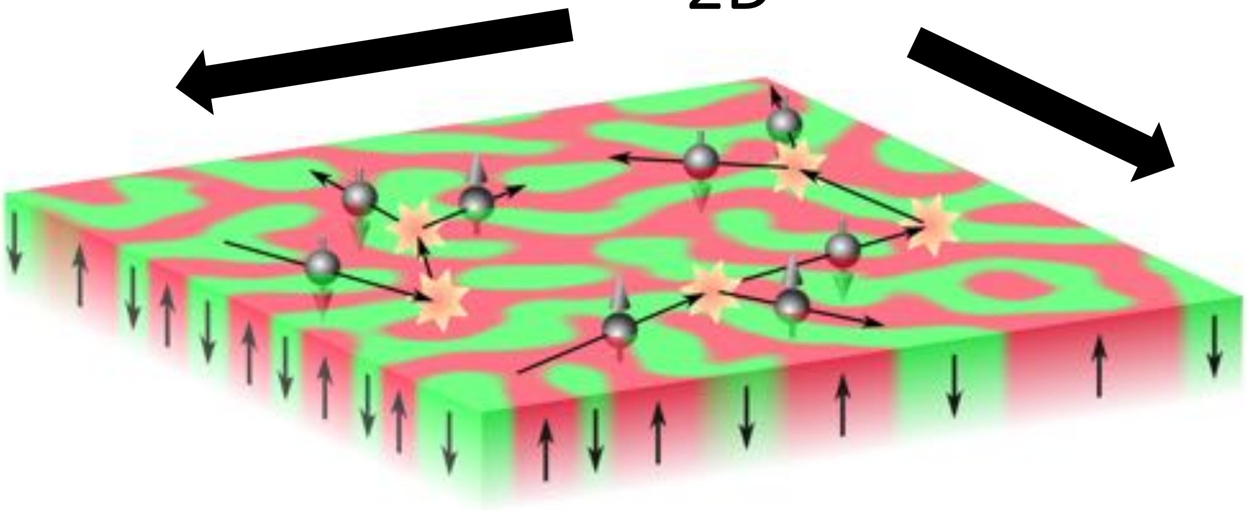
1D



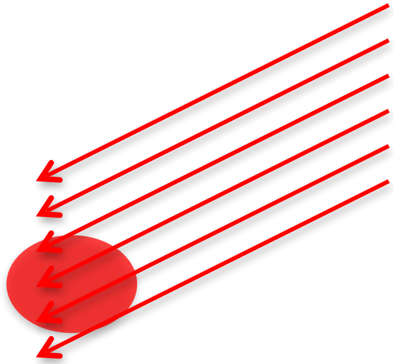
FM - PM

2D

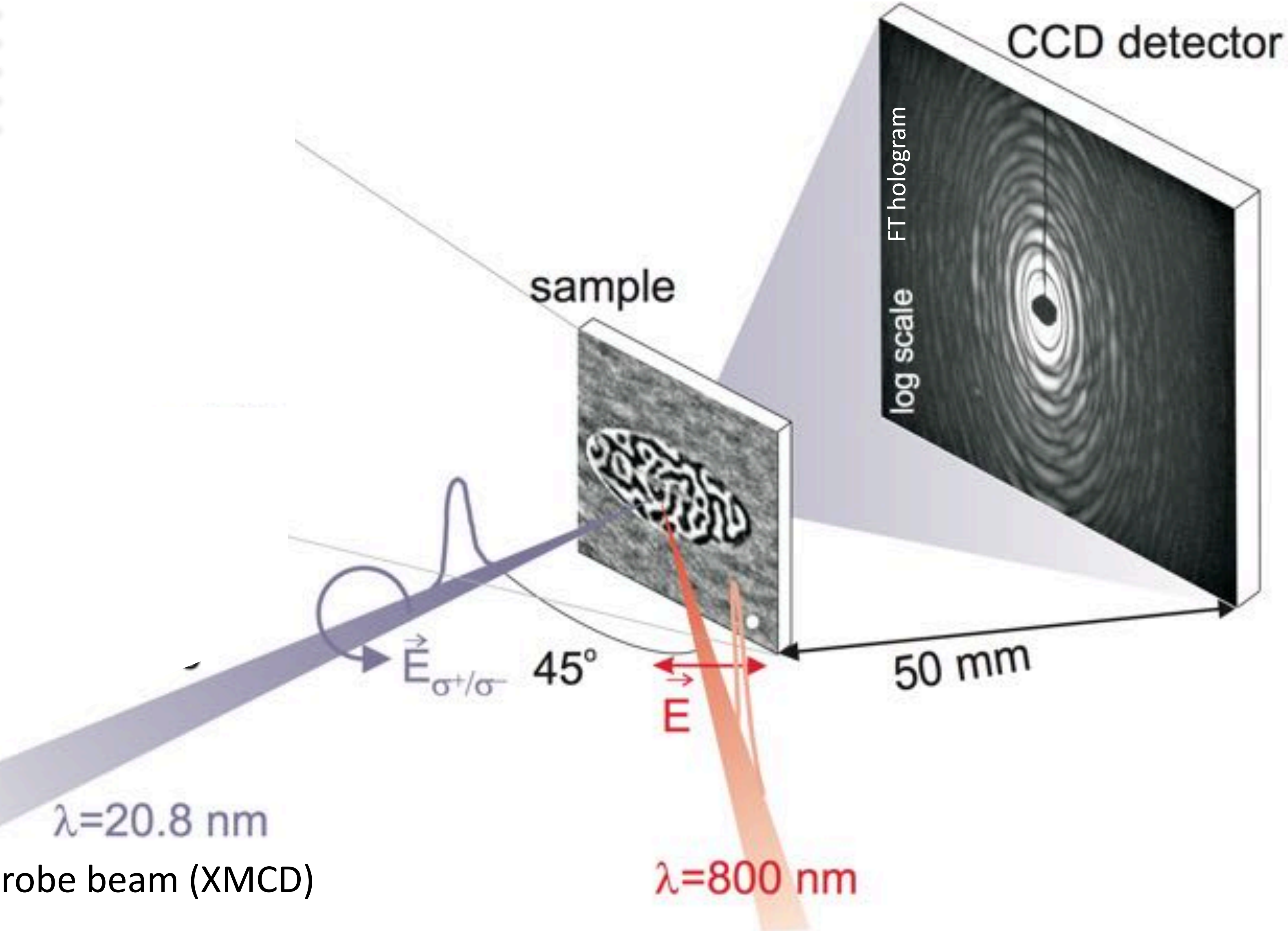


$FM_{\uparrow}$ - $FM_{\downarrow}$

IR-pump X-ray holography probe



E-field enhancement via standing wave



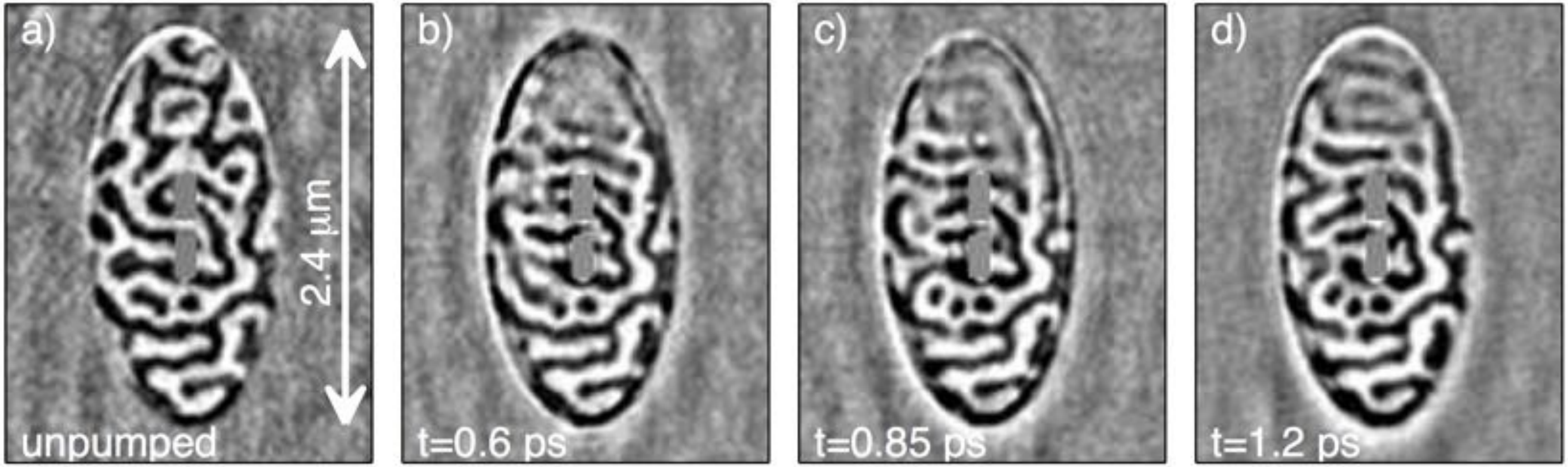
probe beam (XMCD)

pump beam

@ FERMI, DiProI

Seeing demagnetization proceed in time and (real) space

IR-pump FTH-probe @ FERMI



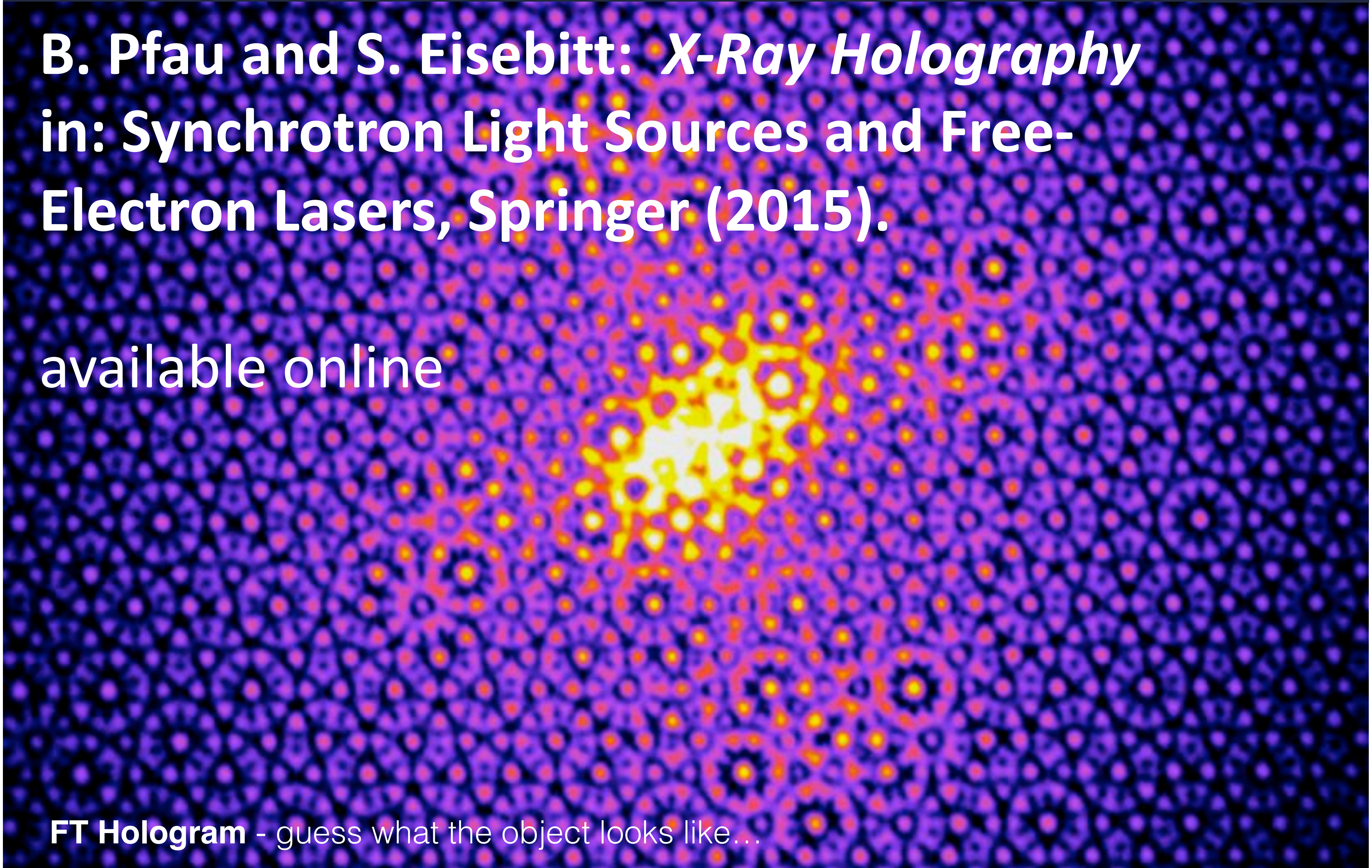
- First ever images of non-local demagnetization (3500 shots per image)
- Demagnetization propagation front moves at 0.2 nm/fs, consistent with spin transport
- Some irreversible changes in domain pattern

C. von Korff Schmising PRL **112**, 217203 (2014)

you want to know details on x-ray holography?

B. Pfau and S. Eisebitt: *X-Ray Holography* in: Synchrotron Light Sources and Free-Electron Lasers, Springer (2015).

available online



FT Hologram - guess what the object looks like…