

# Requirements for Ultrafast Imaging and X-ray Holography

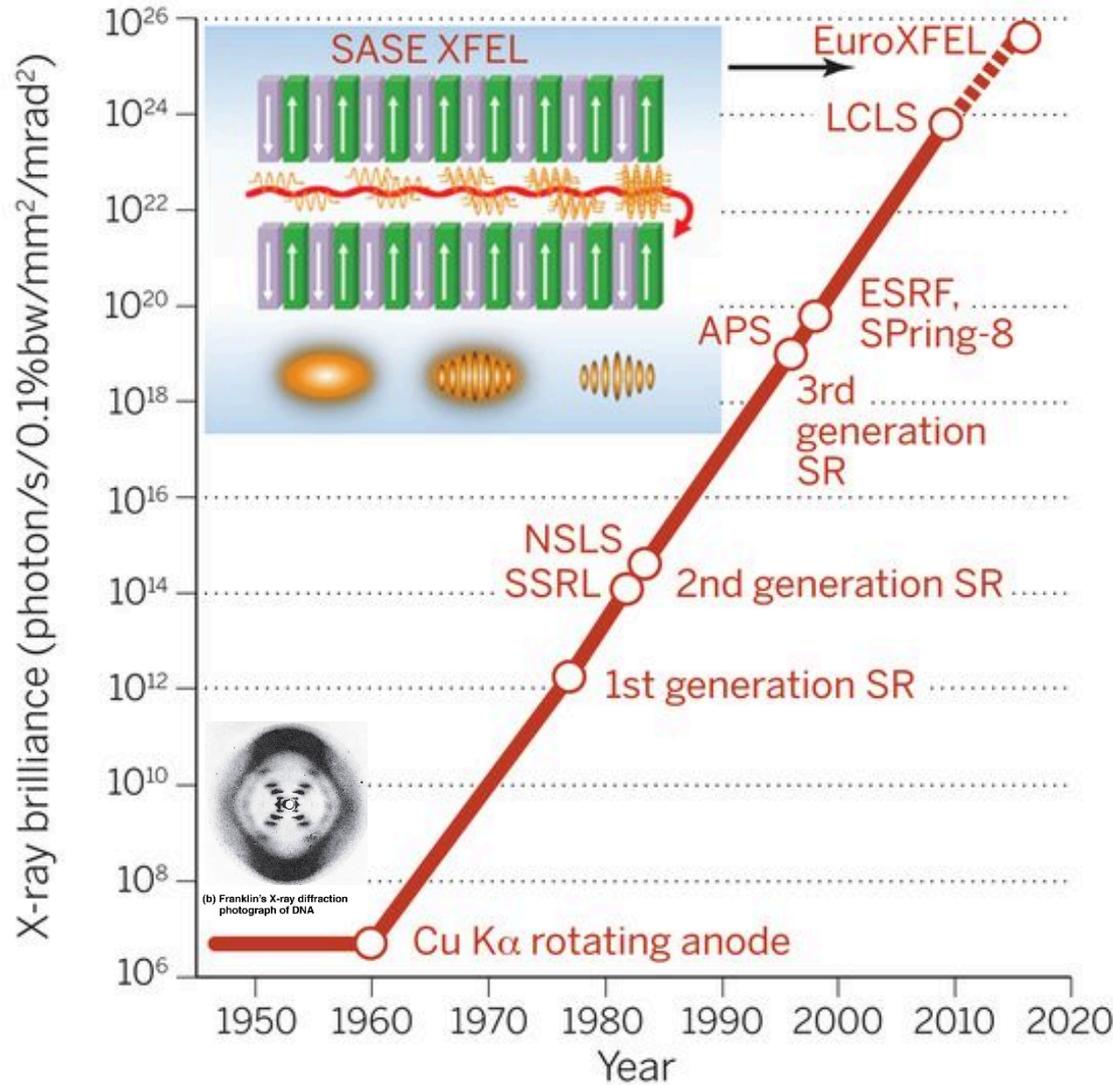
Tais Gorkhover

Stanford PULSE Institute, SLAC



U.S. DEPARTMENT OF  
**ENERGY**

Office of  
Science

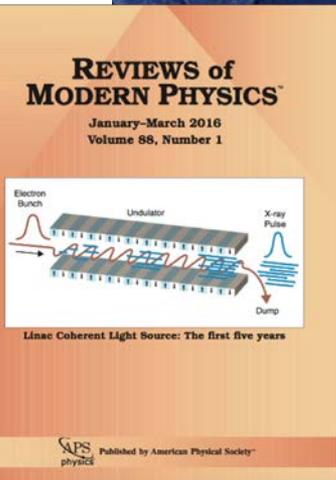
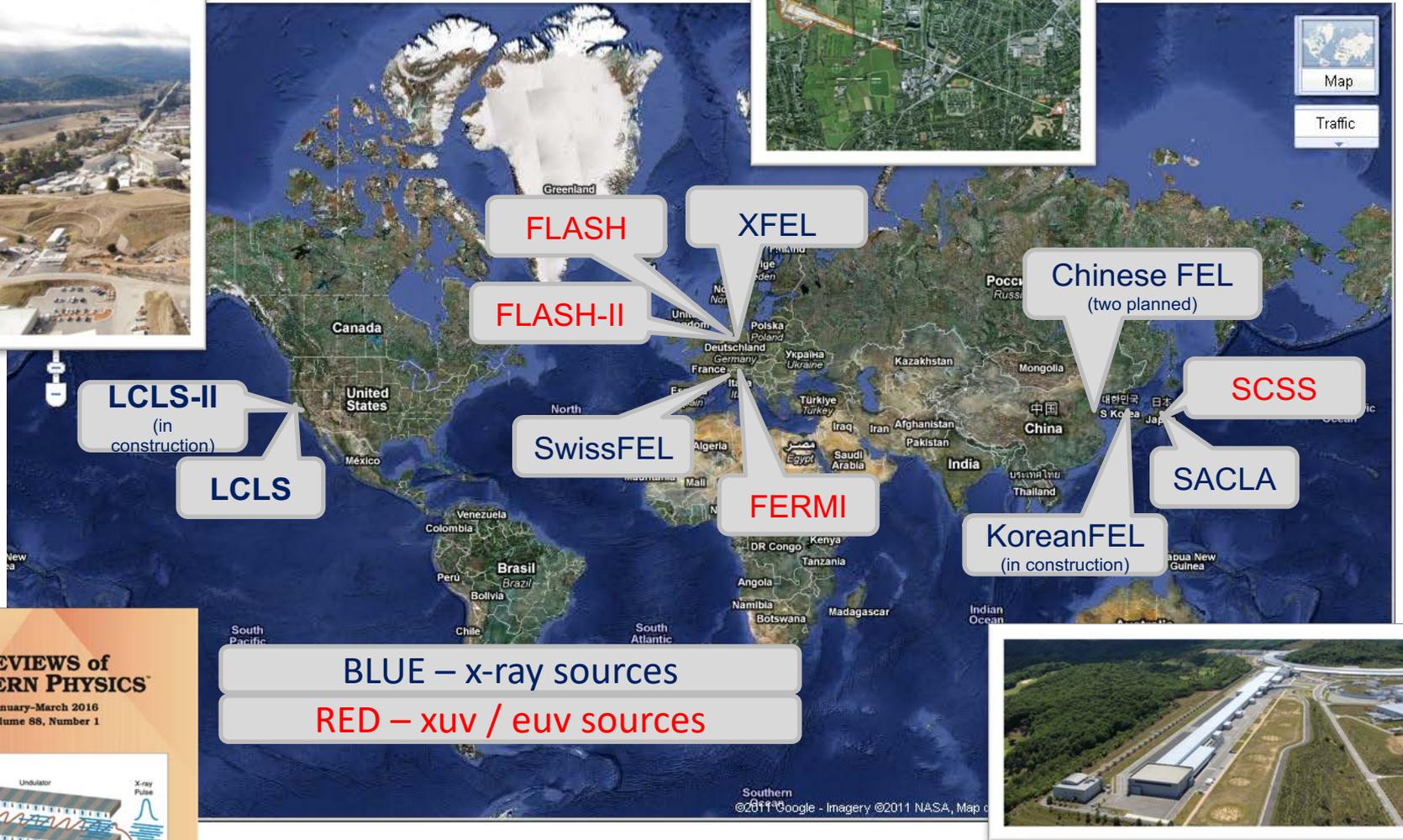
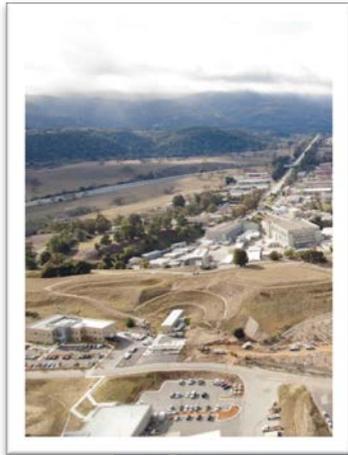


X-ray free electron lasers

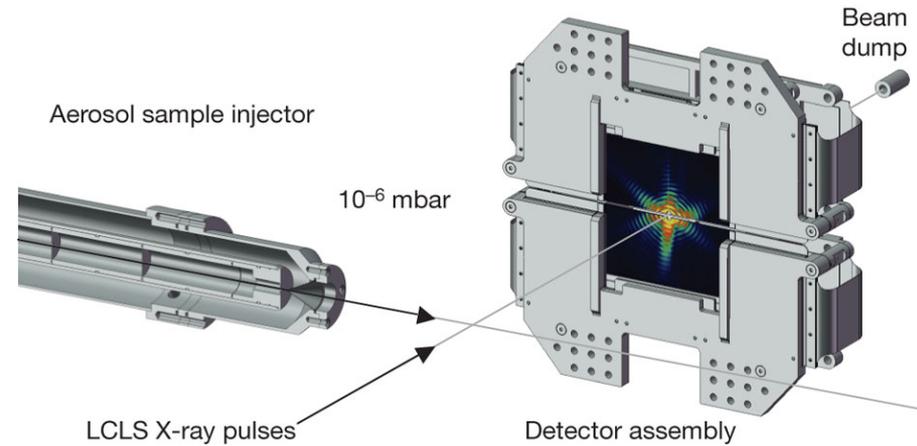
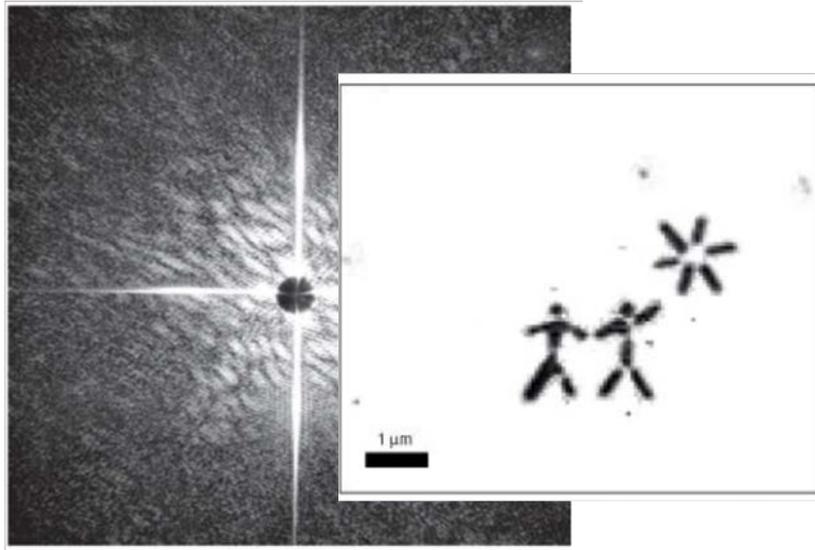
**$10^{11-13}$  X-ray photons in a single, few fs pulse**

are sources of intense X-ray *coherent* radiation

Miao, Jianwei, et al. *Science* 348.6234 (2015): 530-535



Bostedt et al., Rev. Mod. Phys. 88 (2016)

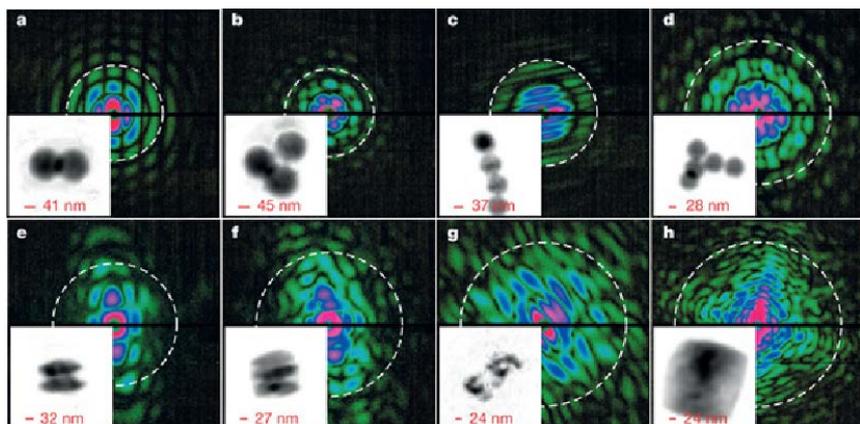


Chapman *et al.* *Nature Physics* **2**, 839–843 (2006)

Seibert *et al.* *Nature* **470**, 78-81 (2011) doi:10.1038/nature09748

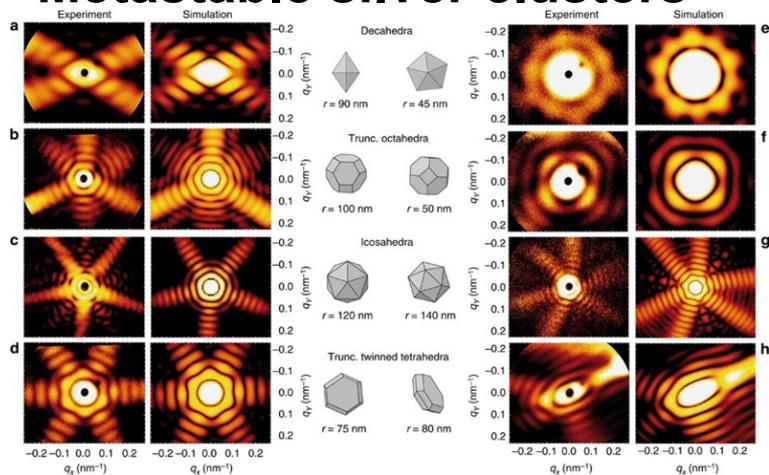
**Potential: High resolution imaging of non-crystalline nanospecimen on the single particle level**

## Soot particles



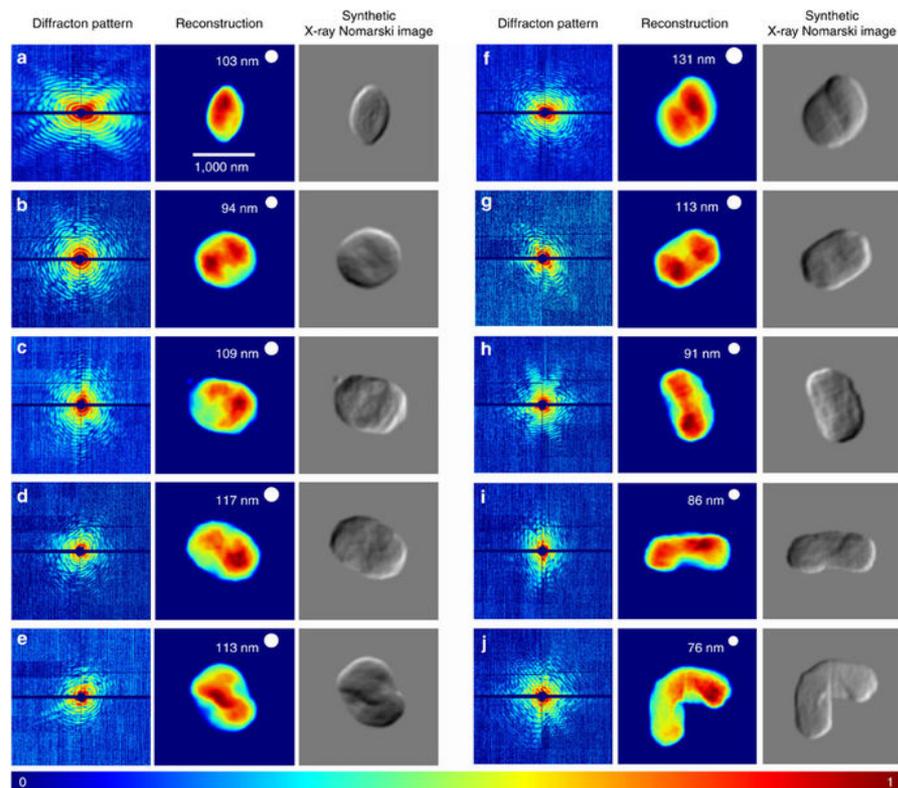
Loh, N. D., et al. *Nature* 486.7404 (2012): 513.

## Metastable silver clusters



Barke, Ingo, et al. *Nat. Comm.* 6 (2015).

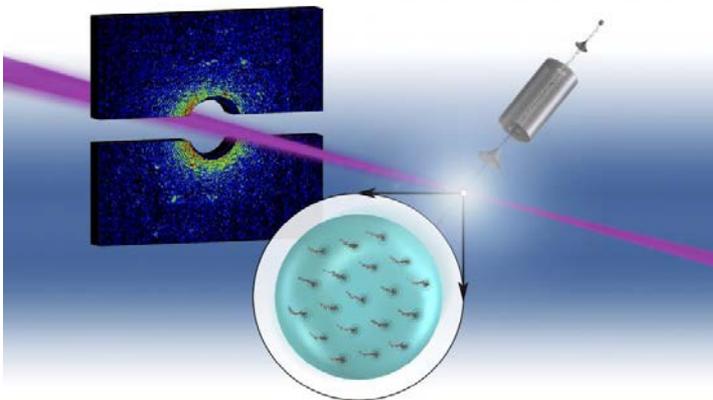
## Living cyanobacteria



van der Schot, *Nat. Comm.* 6:5704 doi: 10.1038/ncomms6704 (2015)

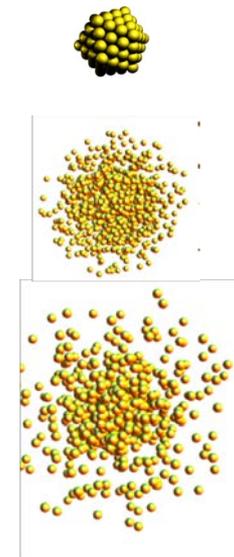
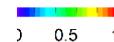
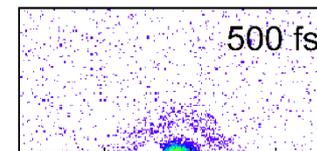
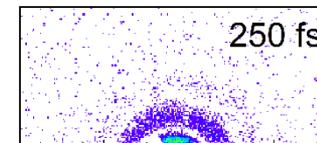
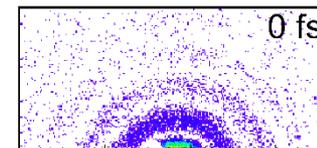
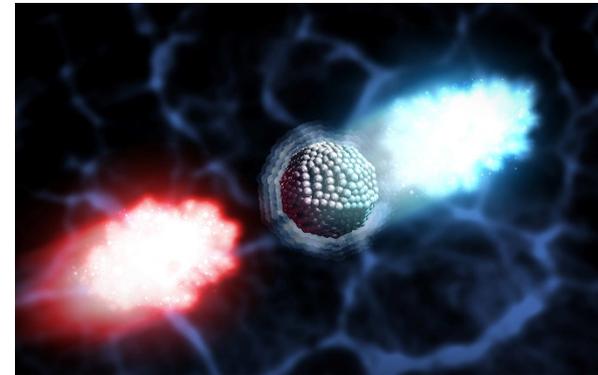
## Ultracold

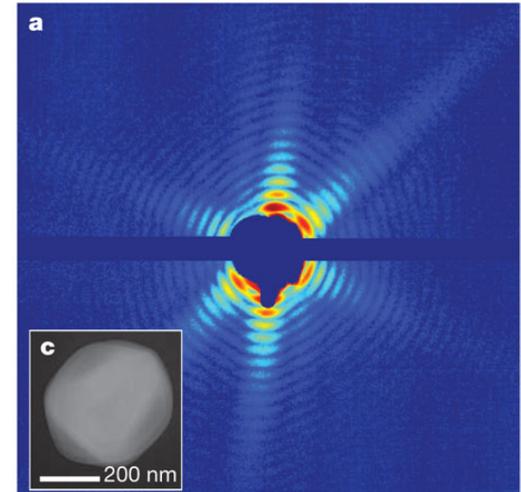
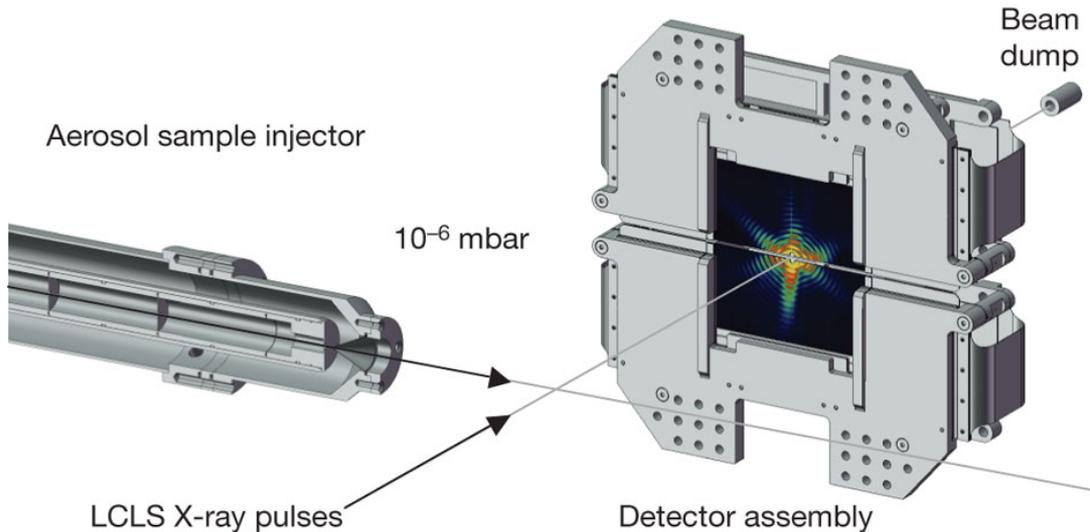
L.Gomez *et al.*, *Science*, 22 August 2014



## Ultrahot

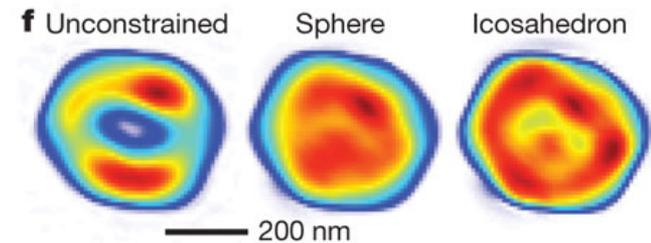
T. Gorkhover *et al.*, *Nature Photonics* 10, (2016)





Seibert *et al.* *Nature* **470**, 78-81 (2011) doi:10.1038/nature09748

**High resolution imaging of non-crystalline nanospecimen on the single particle level**

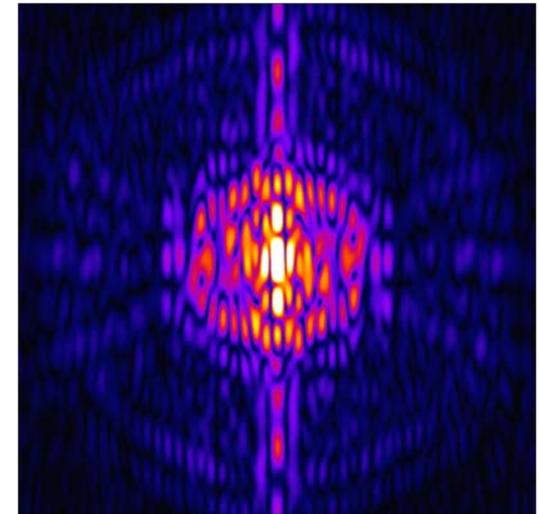


**Phase problem: reconstruction of the structure often not uniquely defined**

# X-ray diffraction imaging

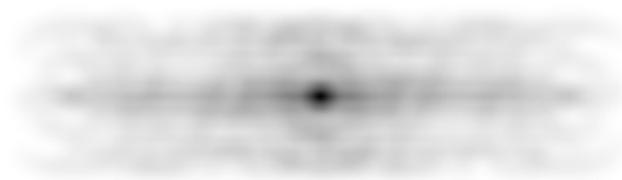


image



reconstruction

Inverse FFT



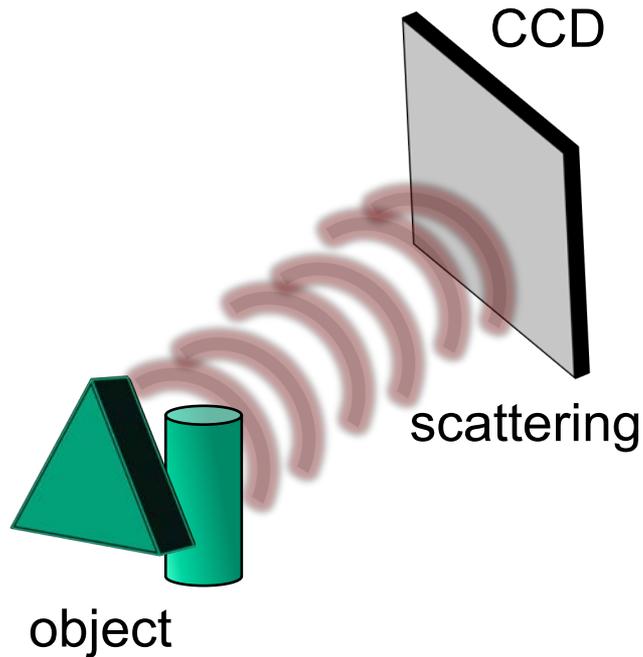
$$I(x, y)$$

$$= |\psi_s(x, y)|^2$$

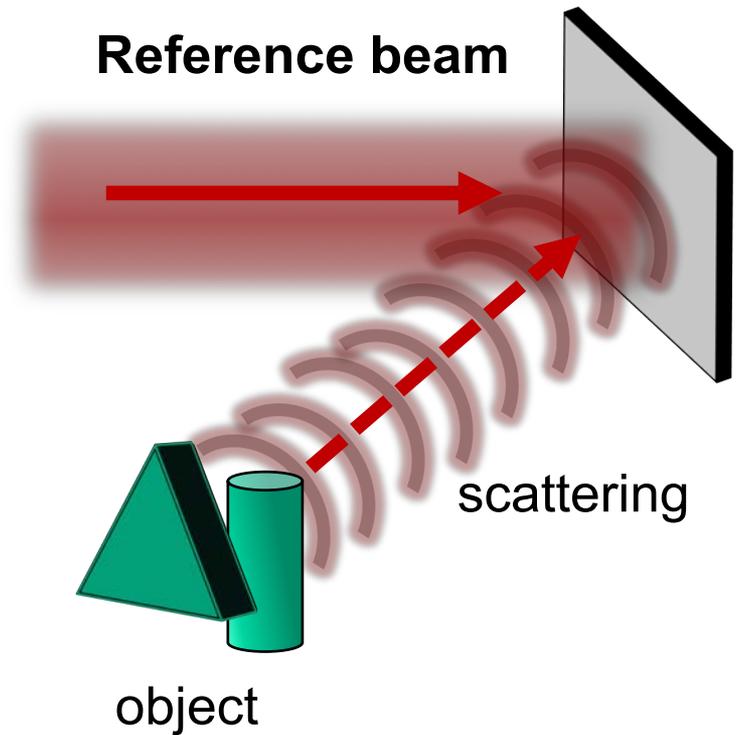
$$= |A_s(x, y)e^{i\varphi_s(x,y)}|^2 = |A_s(x, y)|^2$$

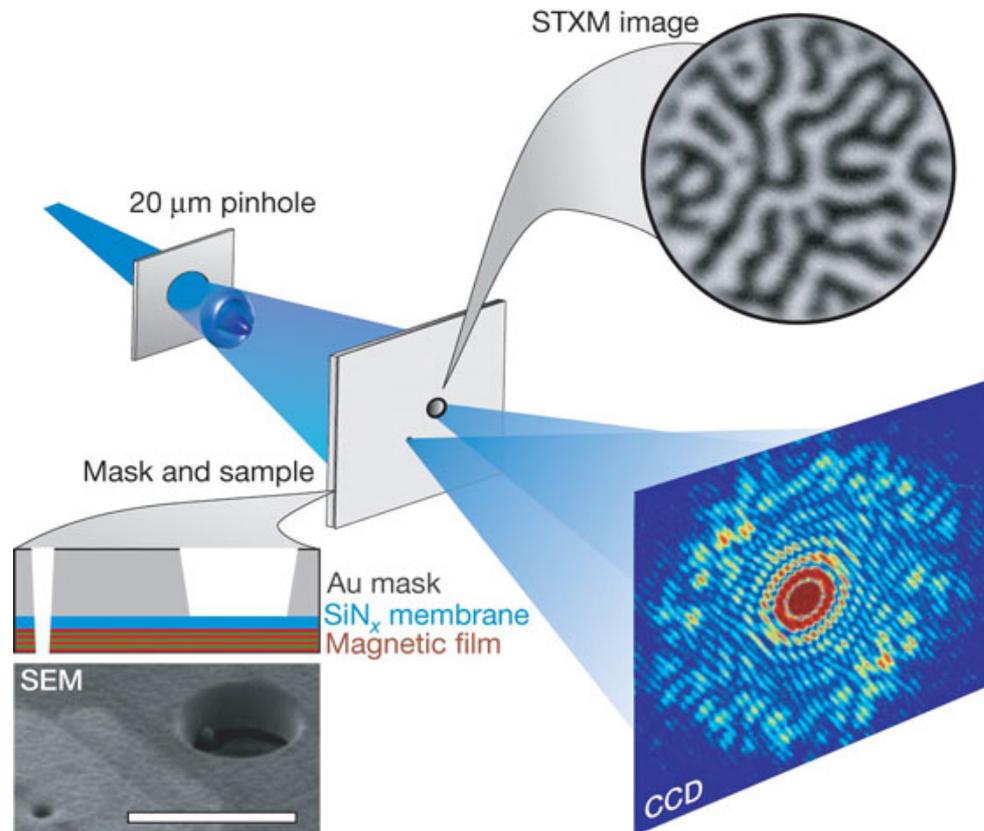
Coherent X-ray diffraction imaging:  
phase information is (almost) lost,  
need for complex iterative  
algorithms

## Coherent imaging diffraction



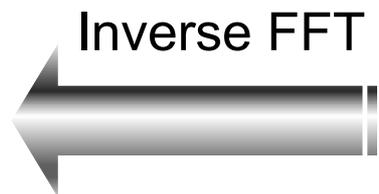
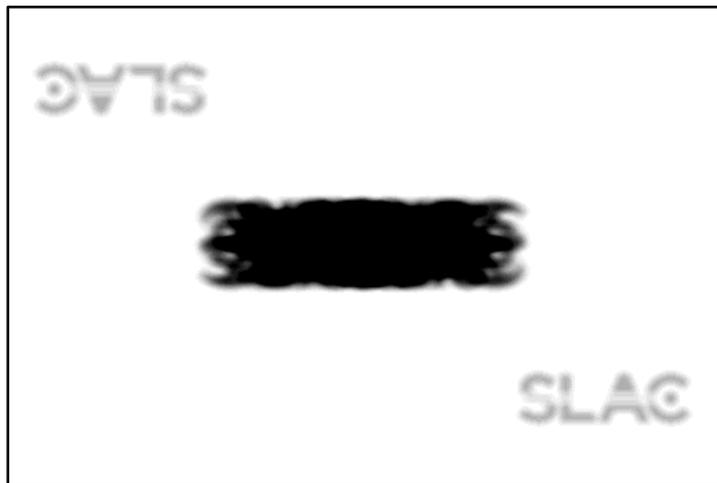
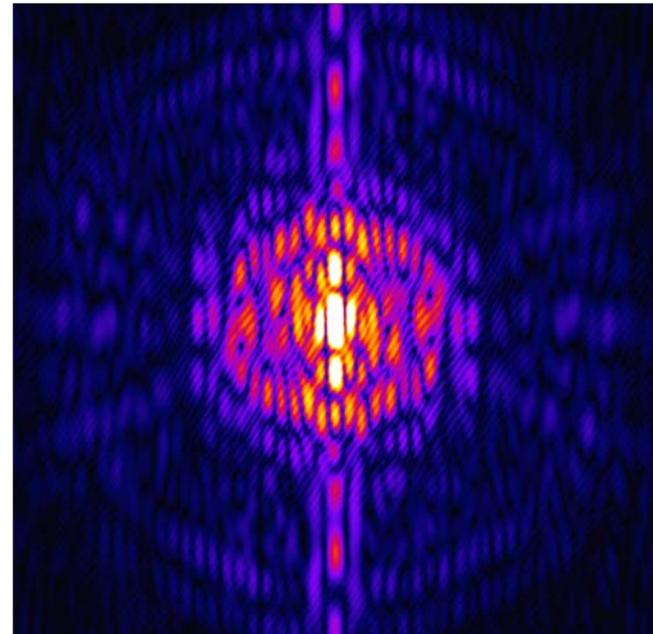
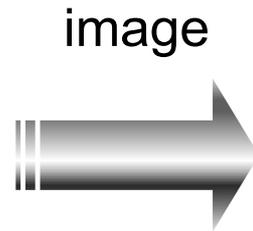
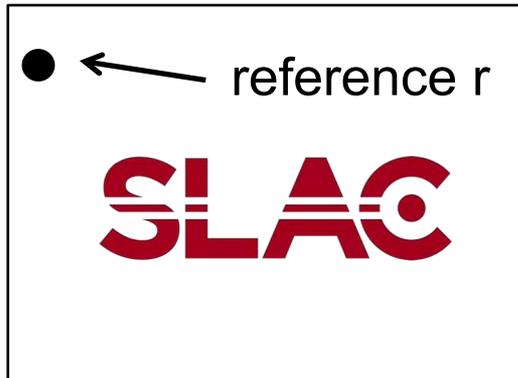
## Holography

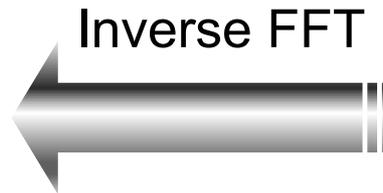
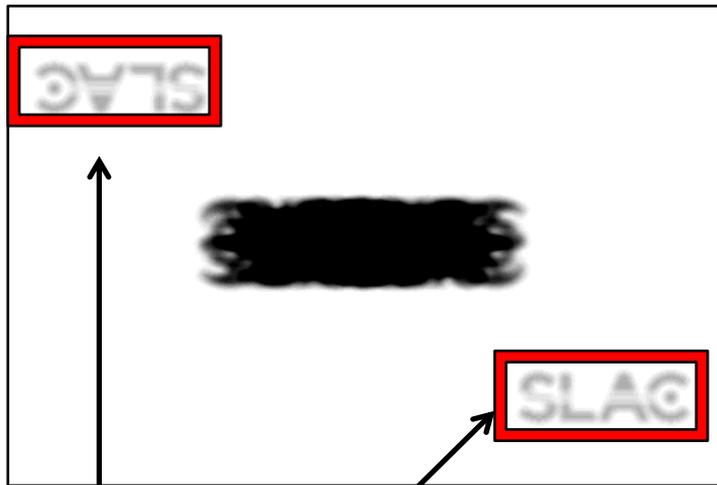
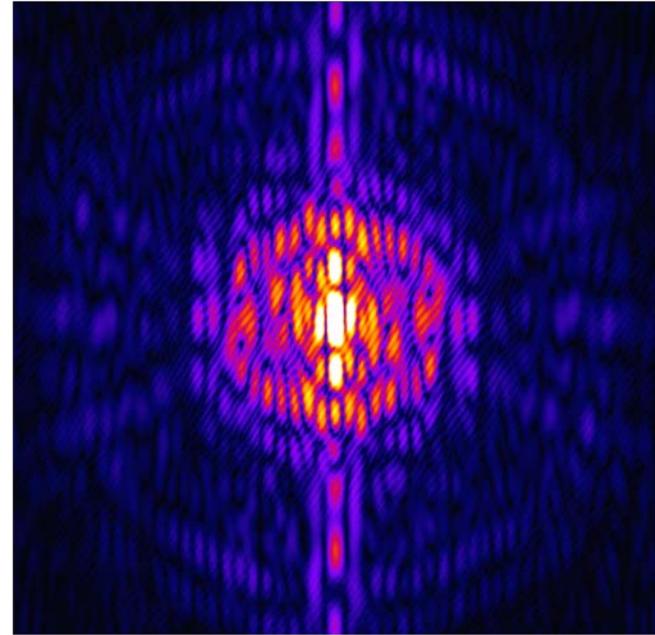
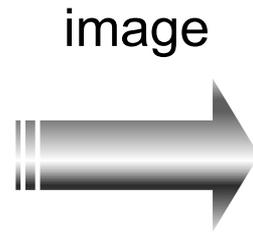
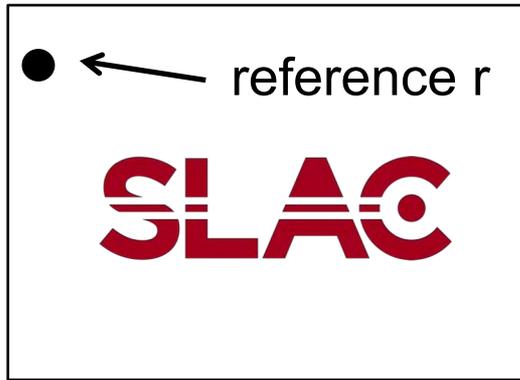




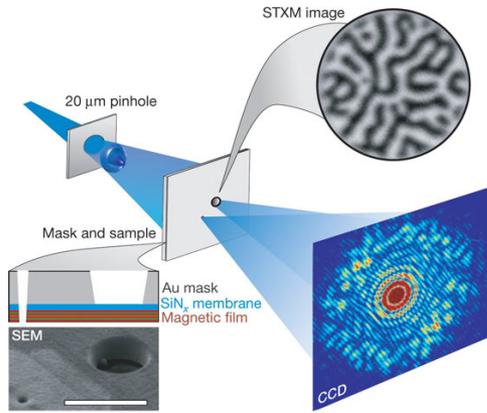
Eisebitt, S., et al., Nature **432**, 885 (2004)

Encoding the phase information directly into the image!





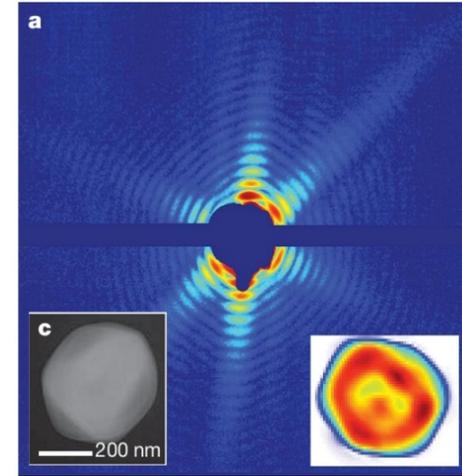
$$\begin{aligned}
 &= |\mathcal{F}\{r + o\}|^2 \\
 &= |R + O|^2 \\
 &= |R|^2 + |O|^2 + \boxed{OR^*} + \boxed{RO^*}
 \end{aligned}$$



Eisebitt, S., et al., *Nature* **432**, 885 (2004)  
Geilhufe, J. et al., *Nature Communications* 5, 3008 (2014)

## X-ray Fourier holography

+



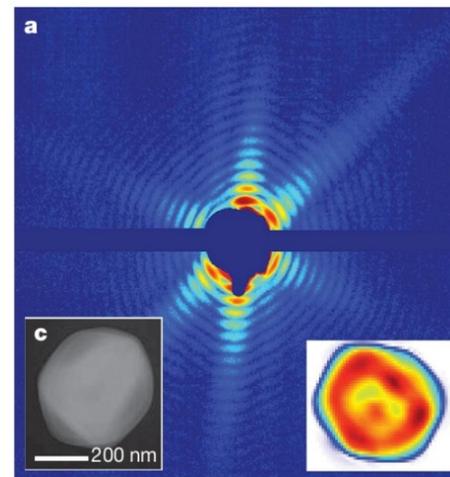
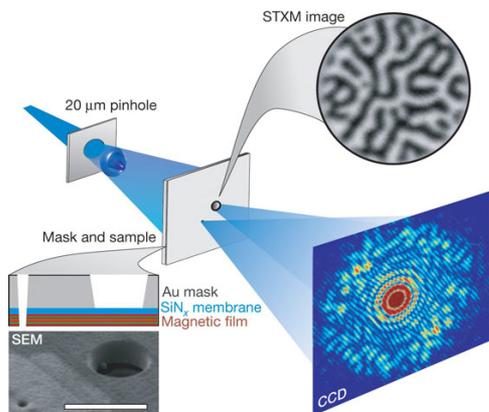
M. M. Seibert et al., *Nature* 470, 78 (2011)

## Single nanoparticle imaging

=

?

# „In-flight“ holography



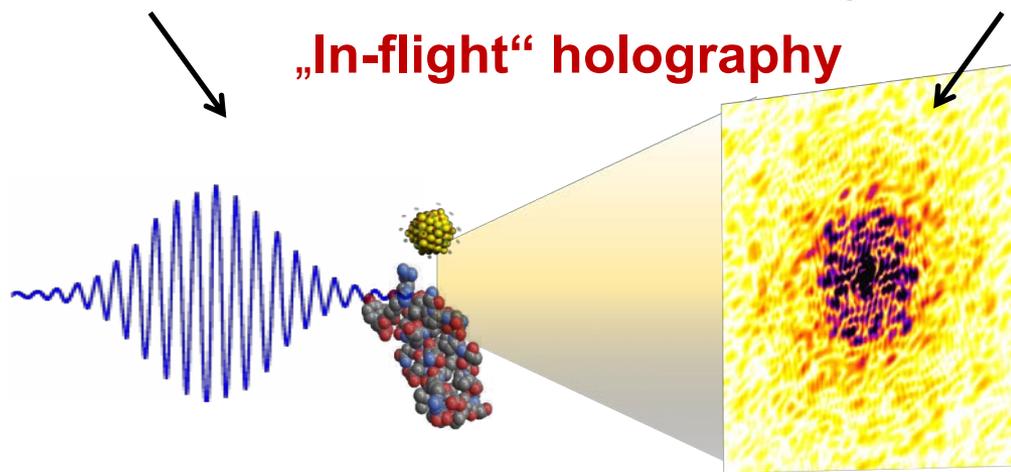
Eisebitt, S., et al., Nature **432**, 885 (2004)

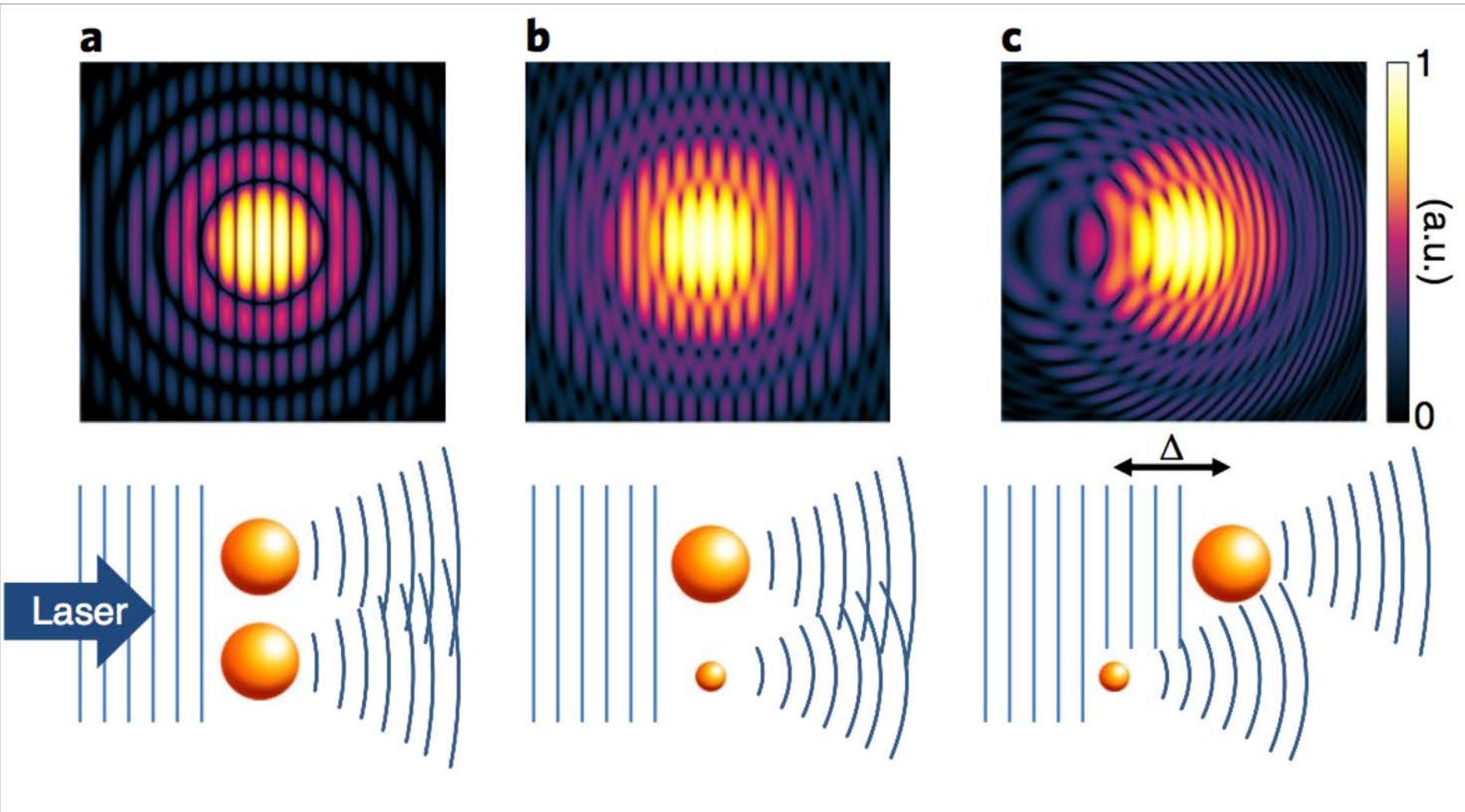
Geilhufe, J. et al., Nature Communications 5, 3008 (2014)

M. M. Seibert et al., Nature 470, 78 (2011)

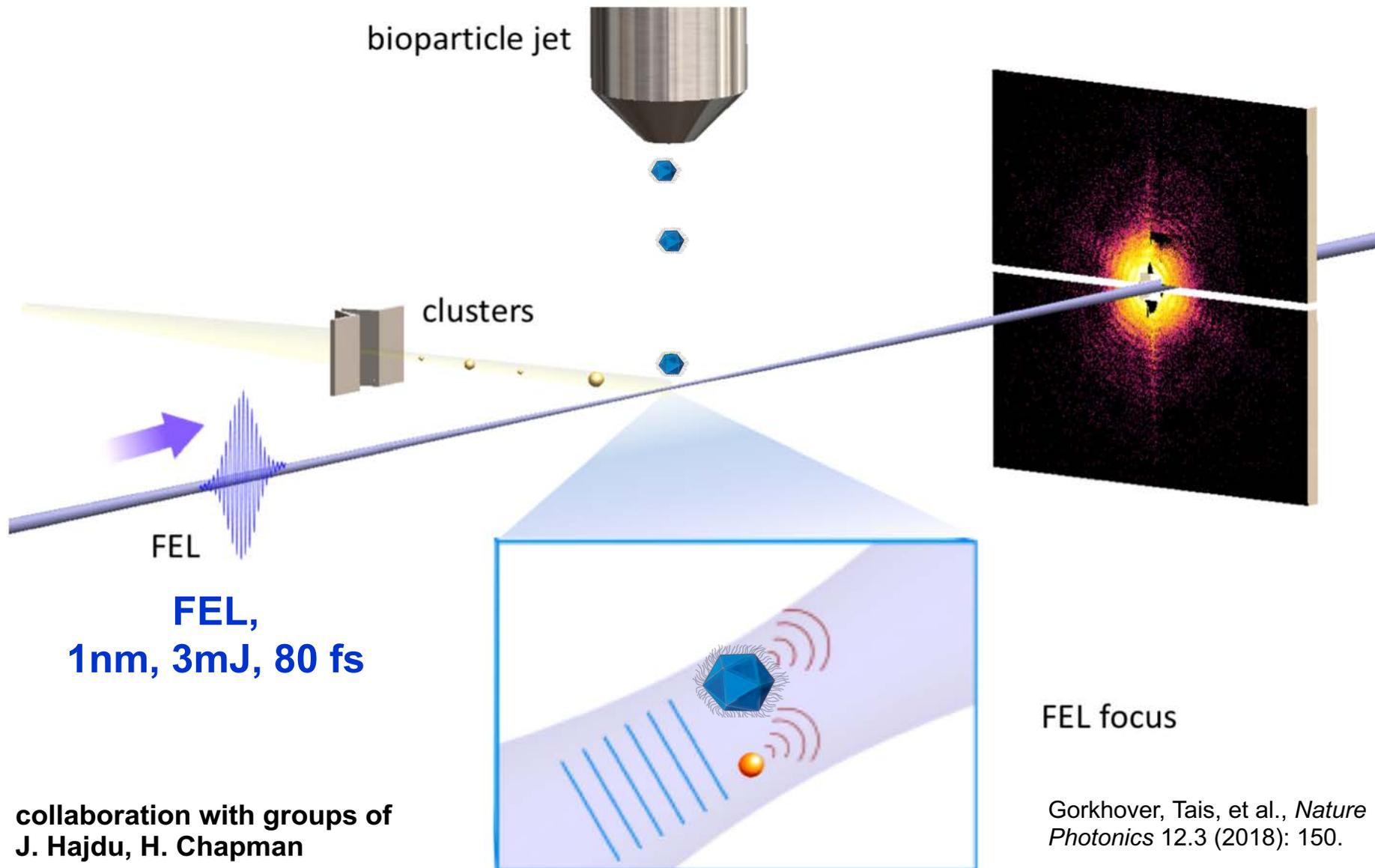
## X-ray Fourier holography

## Single nanoparticle imaging



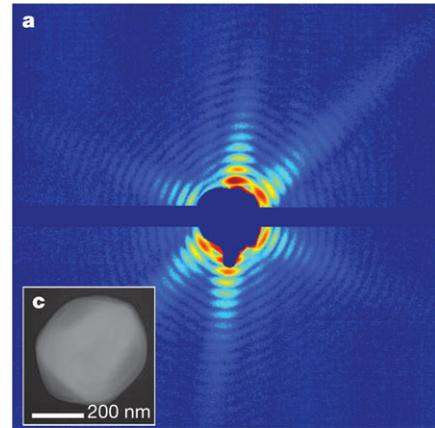


Gorkhover, Tais, et al., *Nature Photonics* 12.3 (2018): 150.

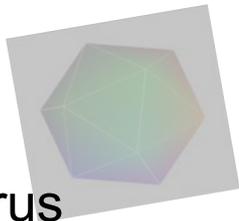


without holography

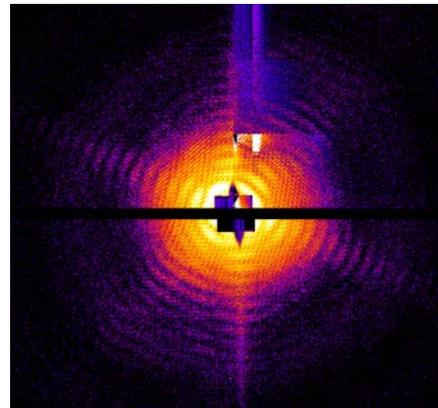
diffraction



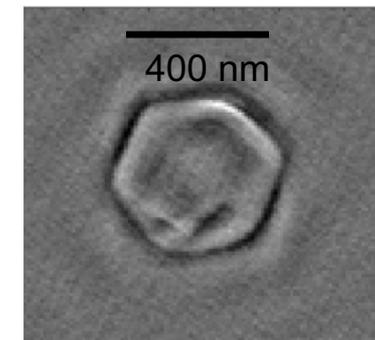
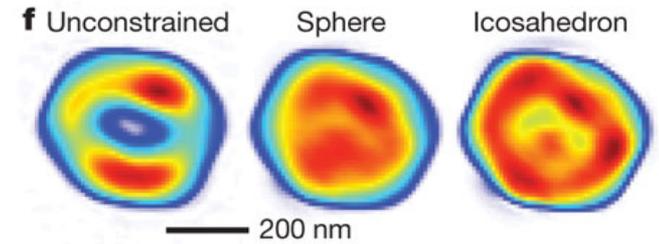
Mimi virus



with holography



reconstruction

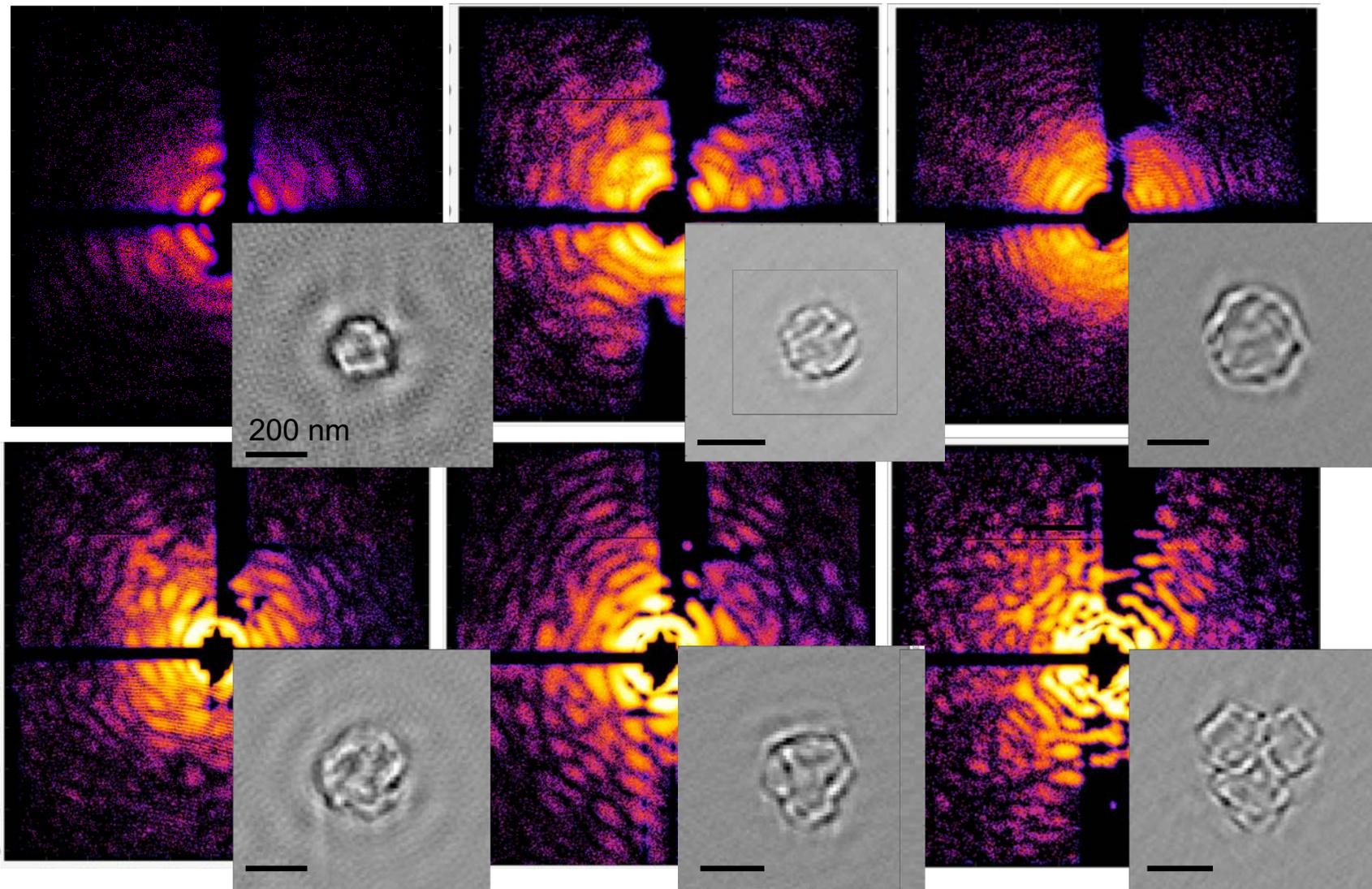


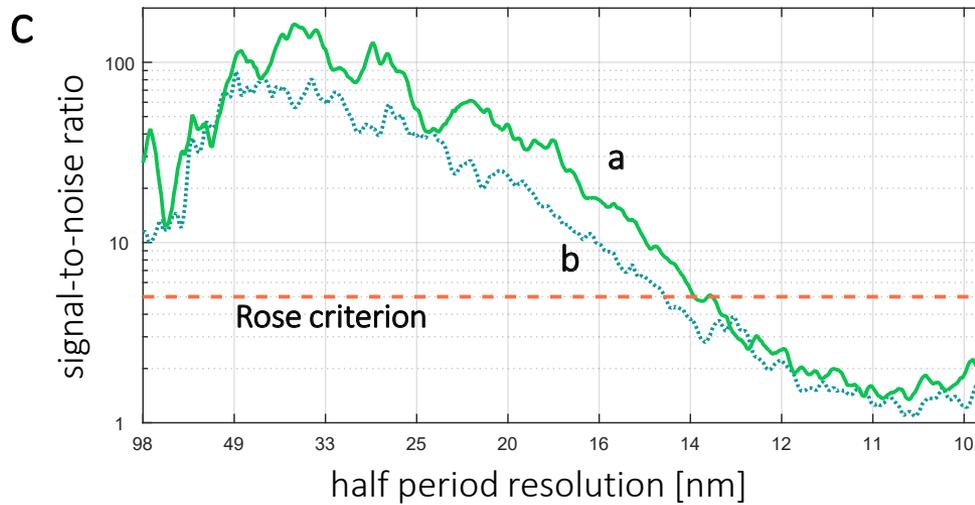
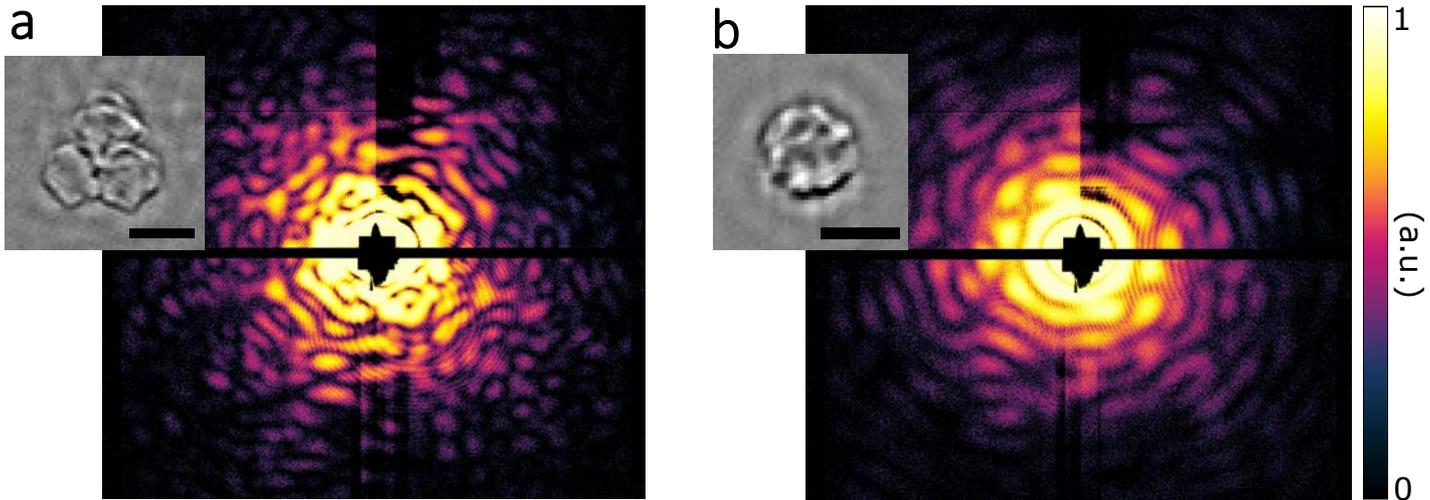
Gorkhover, Tais, et al., *Nature Photonics* 12.3 (2018): 150.

# Inhomogeneous sample set

size →

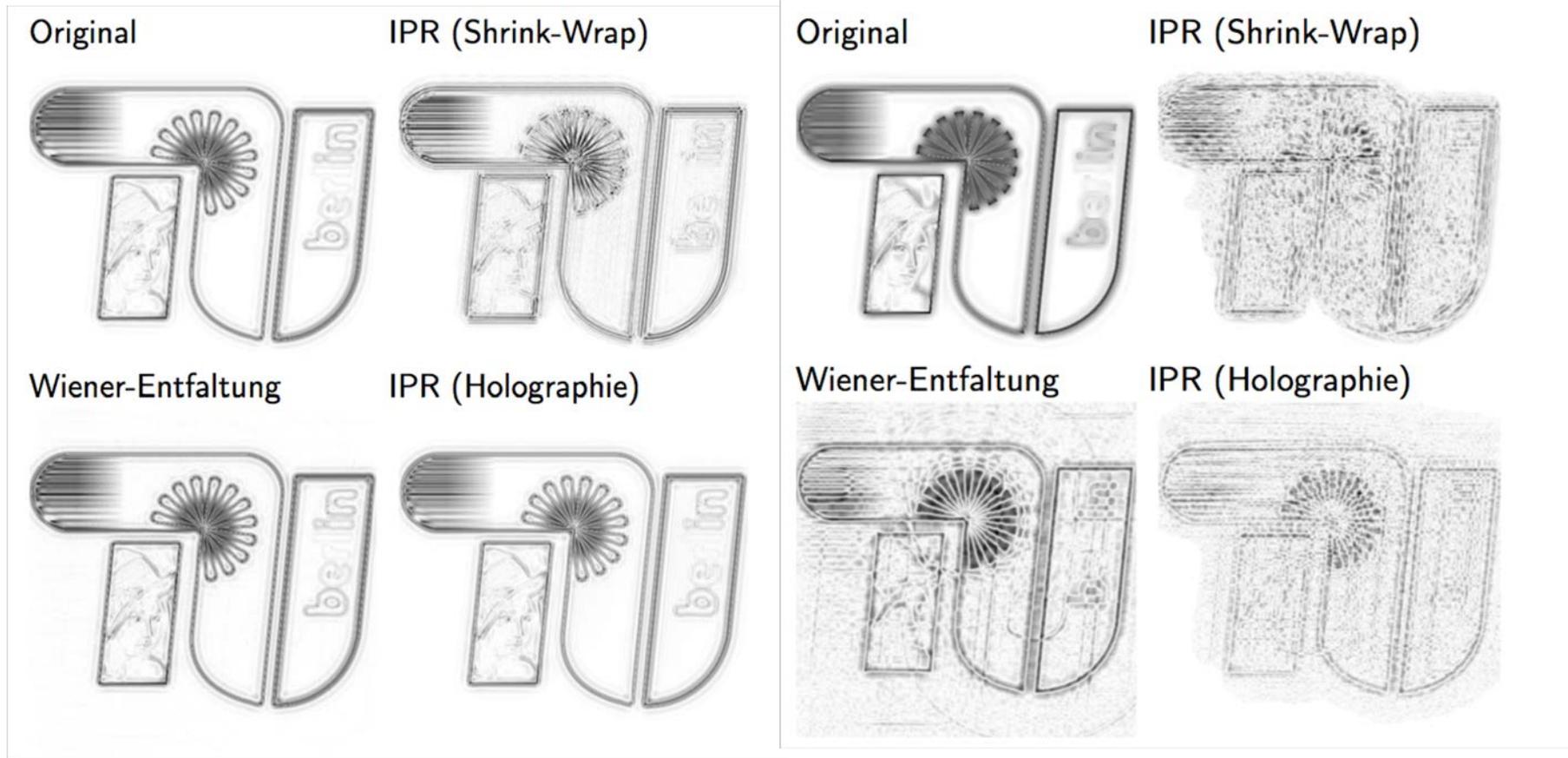
↓ complexity of the image





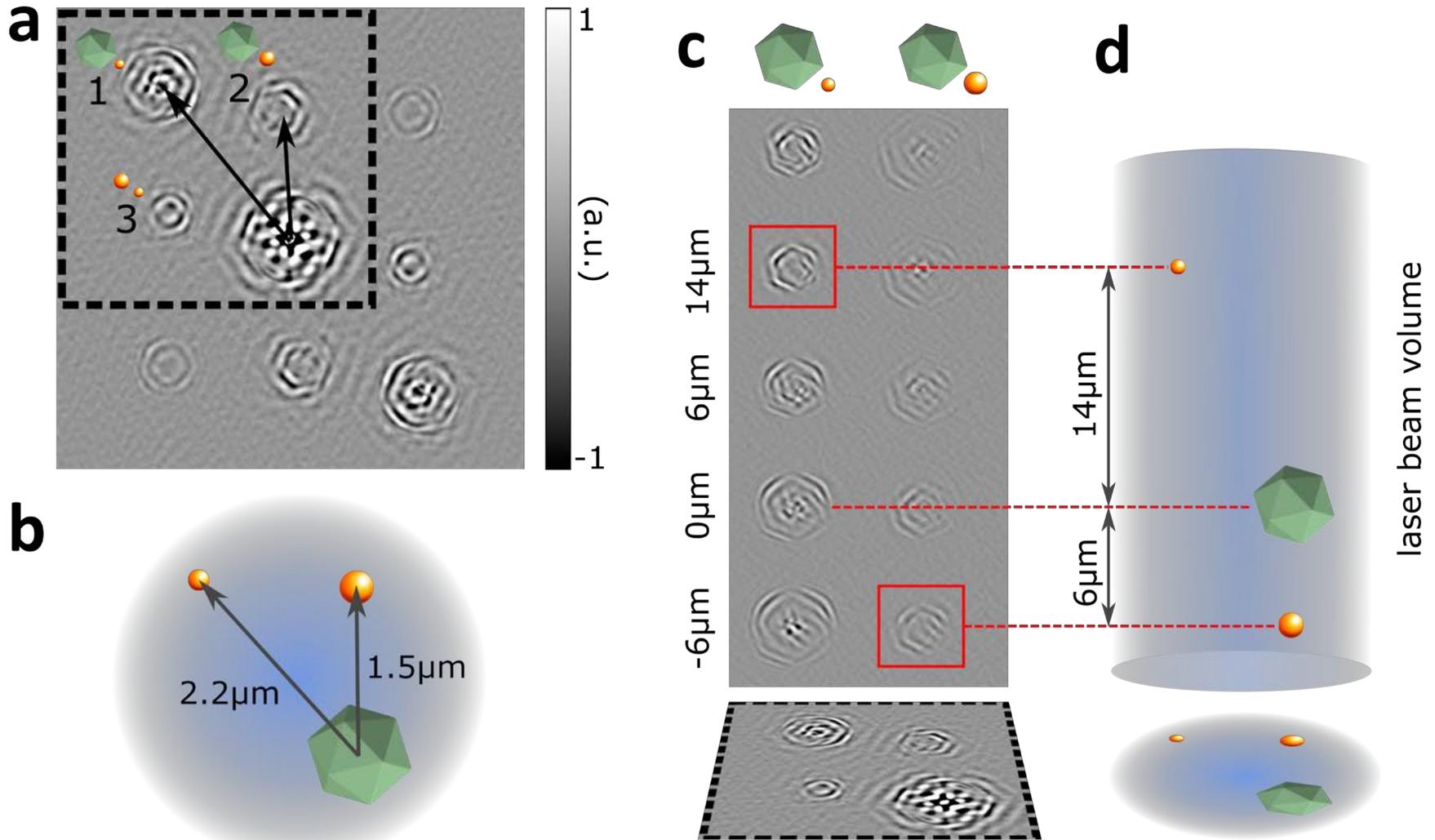
Gorkhover, Tais, et al., *Nature Photonics* 12.3 (2018): 150.

# Holography is robust to noise and imperfections

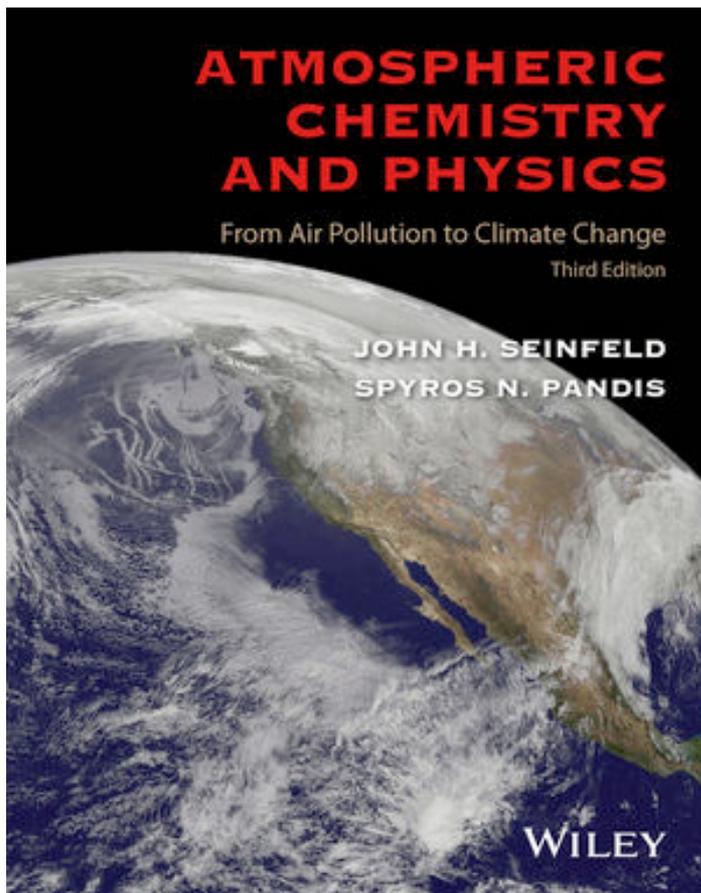


missing information

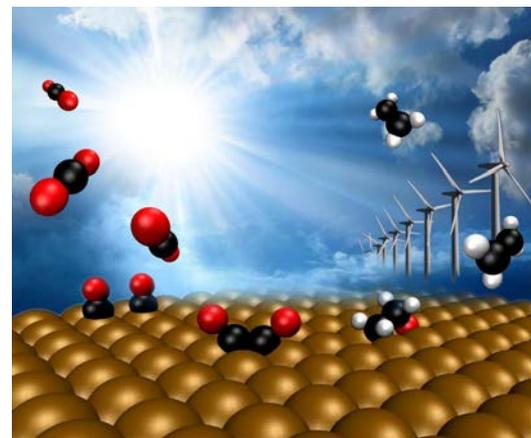
high noise level



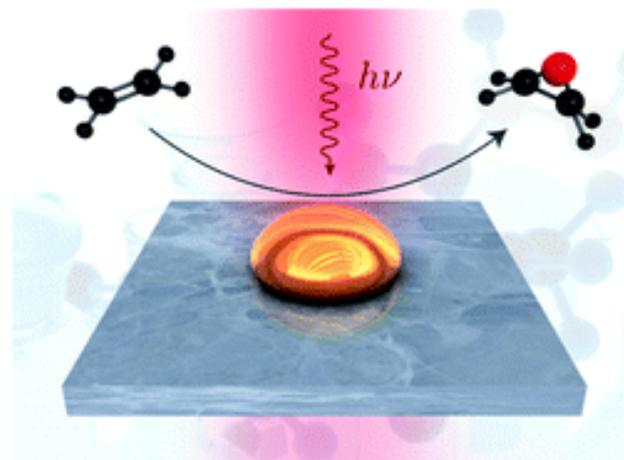
# Application and outlook



**Nucleation dynamics**



**Catalysis**



**Nanoplasmonics**

# Can we reach atomic resolution at single particle level?

These rather high dose densities are difficult to obtain with current and even upcoming x-ray sources. Here, we estimate under which experimental conditions individual atoms can be detected in a molecule or cluster using a single pulse of the XFEL without any additional chemical knowledge. Assuming a fixed coherent dose of about  $D_c = 10^{12}$  photons pulse<sup>-1</sup> for a free-electron laser source, this requires lossless focusing to an area of

$$A = \frac{D_c}{I_c \Delta t}$$

already there

To detect single carbon atoms, the focused area must be smaller than  $A \approx 10 \text{ nm}^2$ . Likewise, for gold atoms, the focused area must be below  $A \approx 2000 \text{ nm}^2$ . For a circular focus, this corresponds to a focus diameter of  $\approx 4 \text{ nm}$  for carbon and  $\approx 50 \text{ nm}$  for gold. For one thing, this limits the

## New Journal of Physics

The open-access journal for physics

Dose requirements for resolving a given feature in an object by coherent x-ray diffraction imaging

Andreas Schropp<sup>1</sup> and Christian G Schroer

Institute of Structural Physics, Technische Universität Dresden, D-01062

Dresden, Germany

E-mail: [schropp@xray-lens.de](mailto:schropp@xray-lens.de)

2018: we can achieve  $d = 60\text{-}80 \text{ nm}$

In theory, we should be able to achieve atomic resolution for heavy atoms metal

2010

# Can we reach atomic resolution at single particle level?

These rather high dose densities are x-ray sources. Here, we estimate under what conditions a single molecule or cluster can be detected in a molecule or cluster using chemical knowledge. Assuming a fixed collection efficiency, this requires loss

$$A = \frac{D_c}{I_c \Delta t}$$

To detect single carbon atoms, the focused area must be below a focus diameter of  $\approx 4$  nm for carbon

## New Journal of Physics

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**Dose requirements for resolving a given feature in an object by coherent x-ray diffraction imaging**

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Dresden, Germany

E-mail: [schropp@xray-lens.de](mailto:schropp@xray-lens.de)

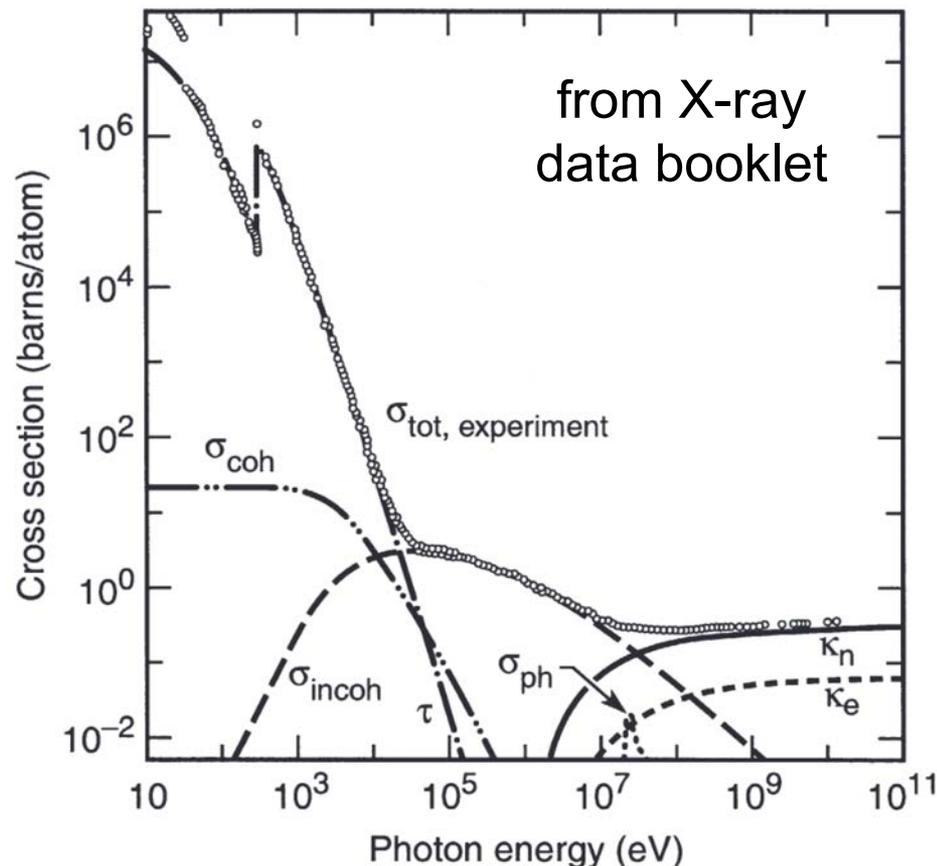
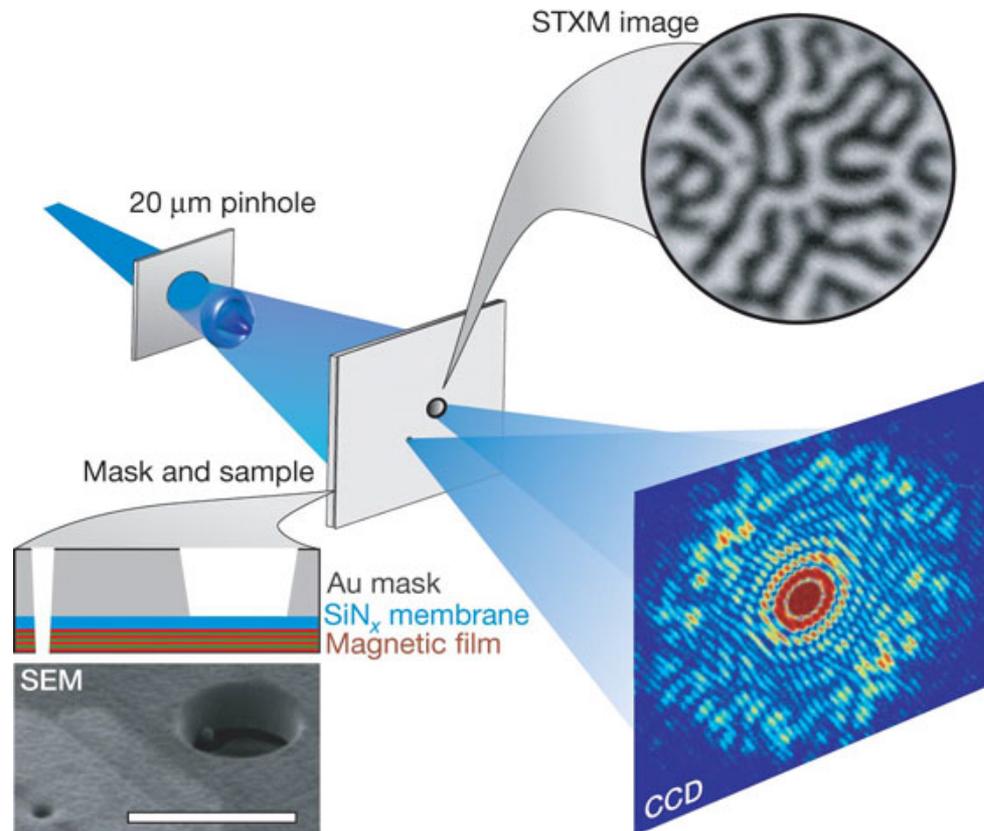
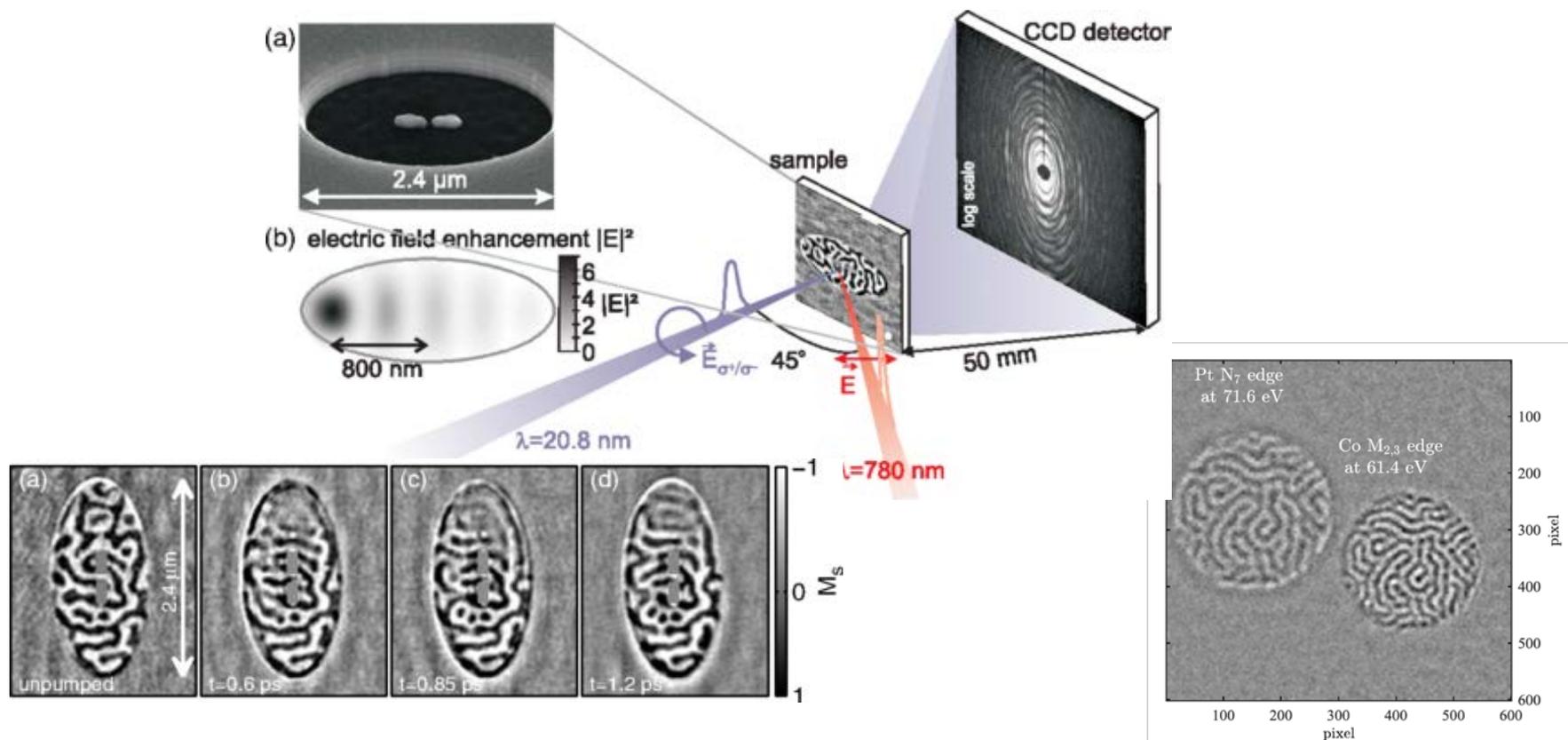


Fig. 3-1. Total photon cross section  $\sigma_{tot}$  in carbon, as a



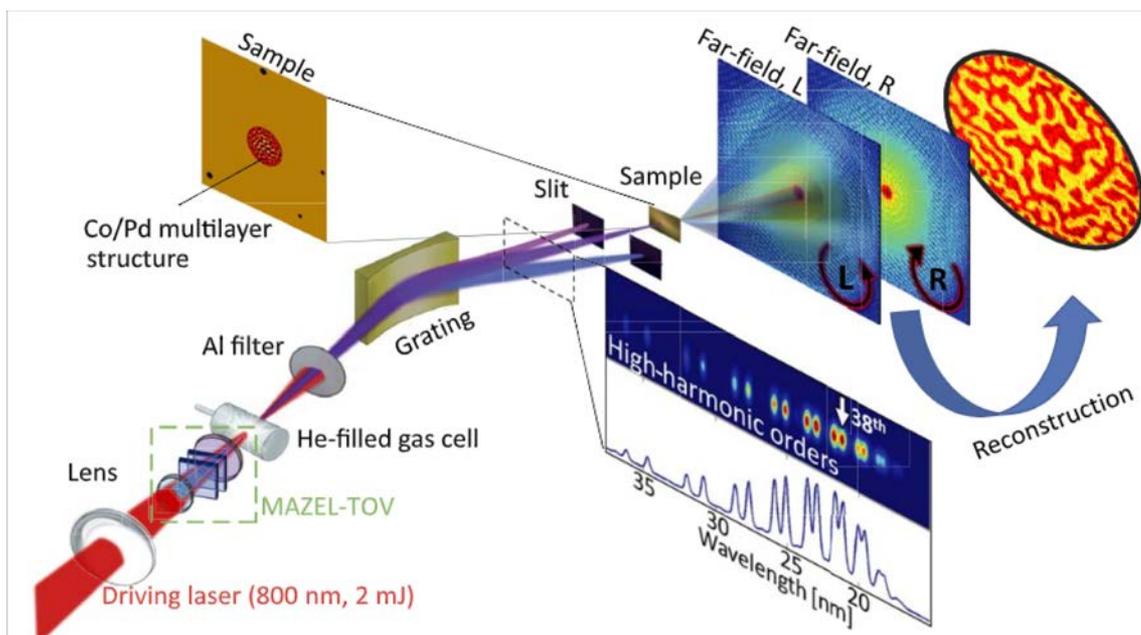
Eisebitt, S., et al., Nature **432**, 885 (2004)

Encoding the phase information directly into the image!

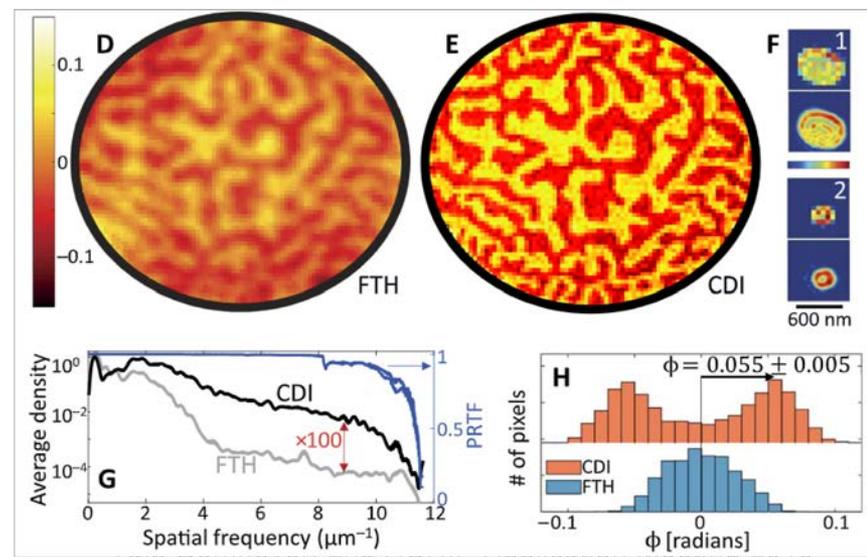


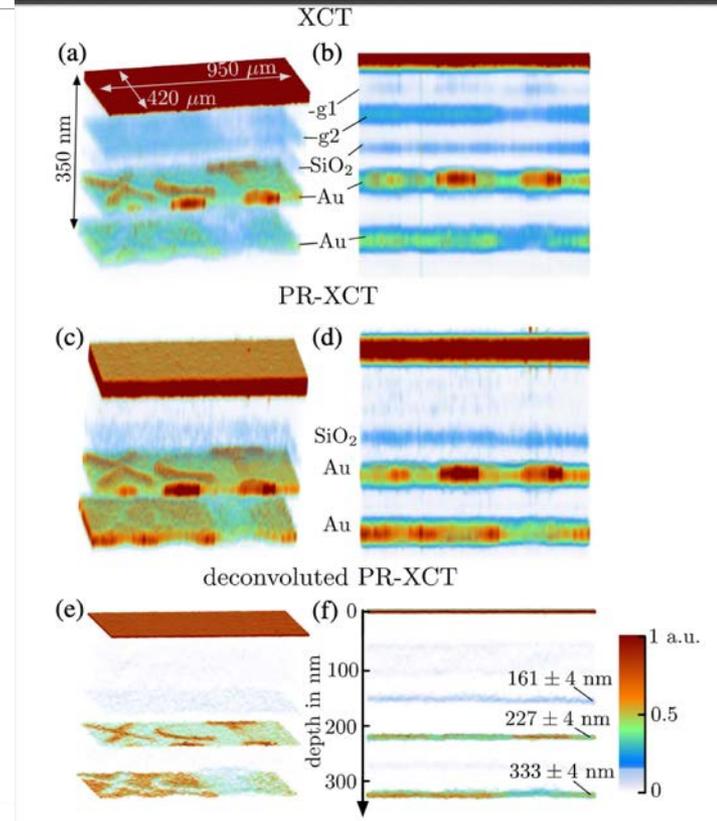
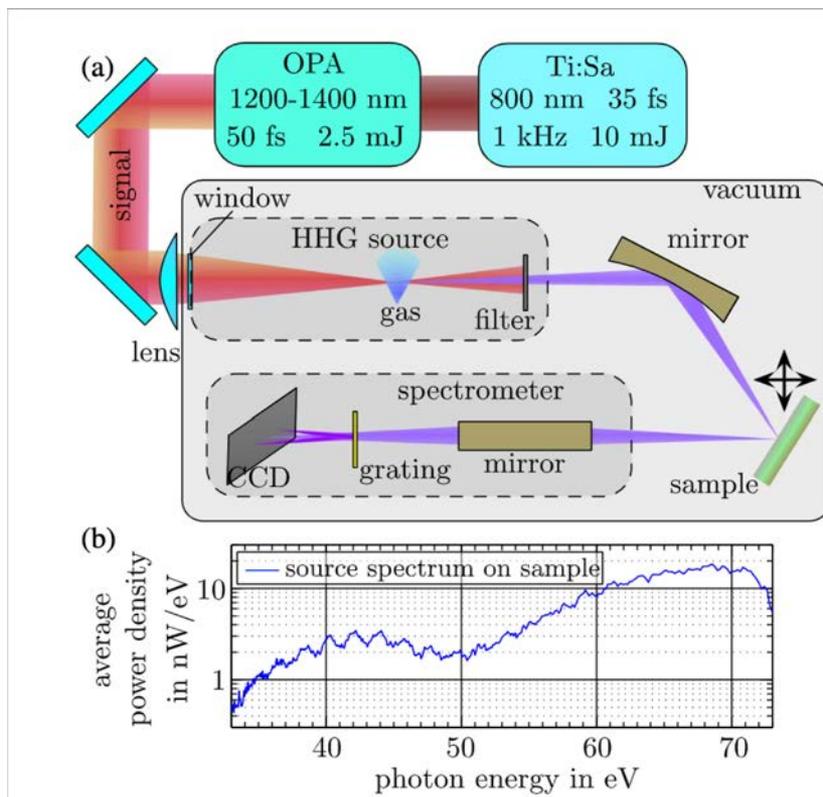
Imaging Ultrafast Demagnetization Dynamics after a Spatially Localized Optical Excitation  
 C. von Korff Schmising, B. Pfau, M. Schneider, C. M. Günther, M. Giovannella, J. Perron, B. Vodungbo, L. Müller, F. Capotondi, E. Pedersoli, N. Mahne, J. Lüning, and S. Eisebitt  
*Phys. Rev. Lett.* **112**, 217203 – Published 29 May 2014

Multi-color imaging of magnetic Co/Pt heterostructures  
 Willems, Felix, Clemens von Korff Schmising, David Weder, Christian M. Günther, Michael Schneider, Bastian Pfau, Sven Meise et al. *Structural Dynamics* 4, no. 1 (2017): 014301.



Kfir, Ofer, et al. "Nanoscale magnetic imaging using circularly polarized high-harmonic radiation." *Science advances* 3.12 (2017): eaao4641.





Fuchs, Silvio, et al. "Optical coherence tomography with nanoscale axial resolution using a laser-driven high-harmonic source." *Optica* 4.8 (2017): 903-906.

## Incoherent Diffractive Imaging via Intensity Correlations of Hard X Rays

Anton Classen,<sup>1,2</sup> Kartik Ayyer,<sup>3</sup> Henry N. Chapman,<sup>3,4,5</sup> Ralf Röhlsberger,<sup>5,6</sup> and Joachim von Zanthier<sup>1,2,\*</sup>

<sup>1</sup>Institut für Optik, Information und Photonik, Universität Erlangen-Nürnberg, 91058 Erlangen, Germany

<sup>2</sup>Erlangen Graduate School in Advanced Optical Technologies (SAOT), Universität Erlangen-Nürnberg, 91052 Erlangen, Germany

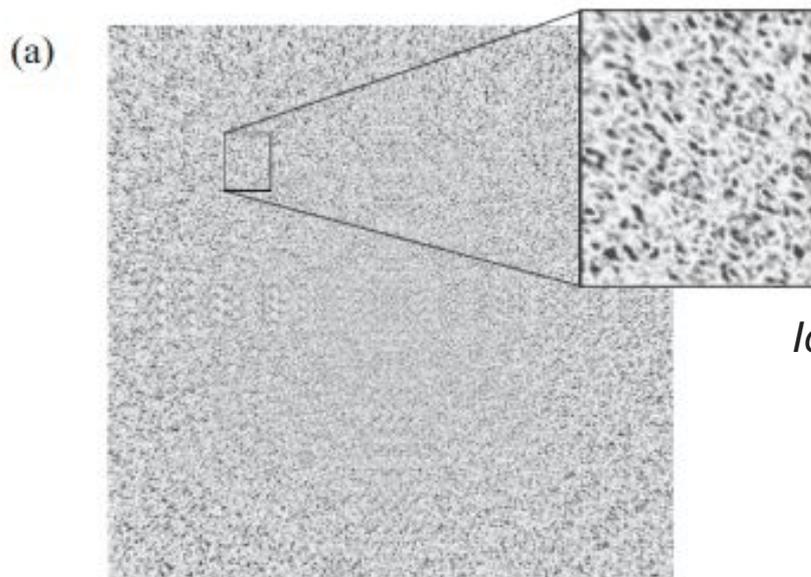
<sup>3</sup>Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany

<sup>4</sup>Department Physik, Universität Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany

<sup>5</sup>The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, 22761 Hamburg, Germany

<sup>6</sup>Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany

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*Ideal for X-ray pulses < 1 fs*

