Flash x-ray holography

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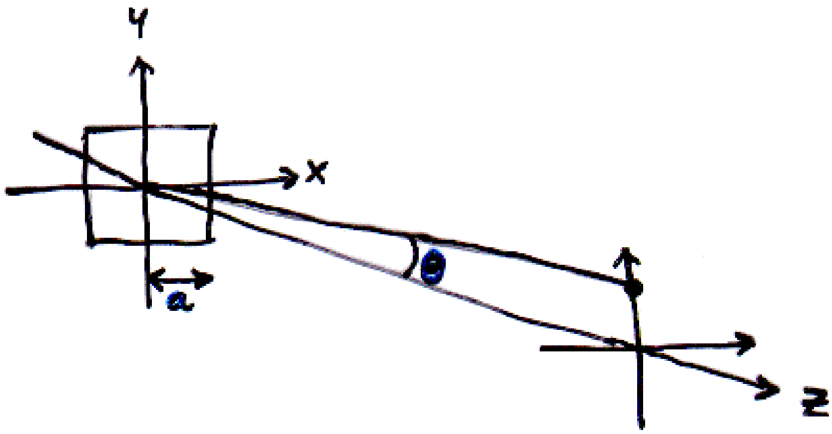
Advanced Photon Source

Why x-ray holography?

- Multiple-view 3D imaging at low NA
- Single-view 3D imaging at high NA
- Elemental and chemical specificity
- Phase as well as absorption contrast
- Can be coupled with interferometric methods
- Simple optical geometries required
- Parallel image acquisition → array detectors
- Suited to flash sources → avoid radiation damage

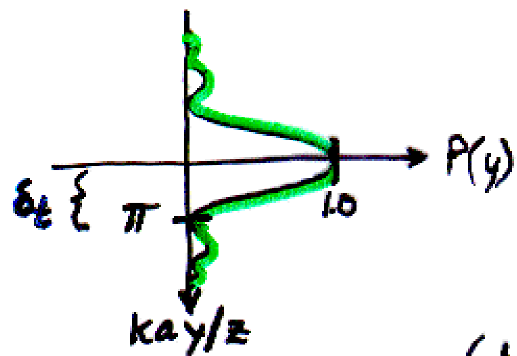
Imaging Resolution



Point-Spread Function

$$P(x,y) = \frac{\sin X}{X} \cdot \frac{\sin Y}{Y}$$

$$X = \frac{kax}{z} \qquad Y = \frac{kay}{z}$$



$$\Rightarrow \boxed{\delta_t = 0.5 \frac{\lambda}{NA}}$$ transverse

where $\begin{cases} NA \sim \frac{a}{z} \\ k = \frac{2\pi}{\lambda} \end{cases}$

$$\vec{\Delta k} = \vec{K}_{inc} - \vec{K}_{scatt} \quad ; \quad 0 \leq \Delta k \leq 2K_{inc}$$

↑ (180°)

Elastic (Coherent) Scattering

$$k_x^2 + k_y^2 + (k_z + k_{inc})^2 = k_{inc}^2$$

For extreme-angle ray (yz plane, $k_x = 0$) with $k_y = \frac{2\pi}{2\delta_t} = \frac{2\pi}{\lambda} NA$

$$k_z = \frac{k_y^2}{2k_{inc}} \qquad (k_z \ll K_{inc})$$

$$\Rightarrow \boxed{\delta_l = \frac{\lambda}{(NA)^2}}$$ longitudinal

Two methods, two regimes

- **External reference**

  Use x-ray optics to form reference wave and object illumination

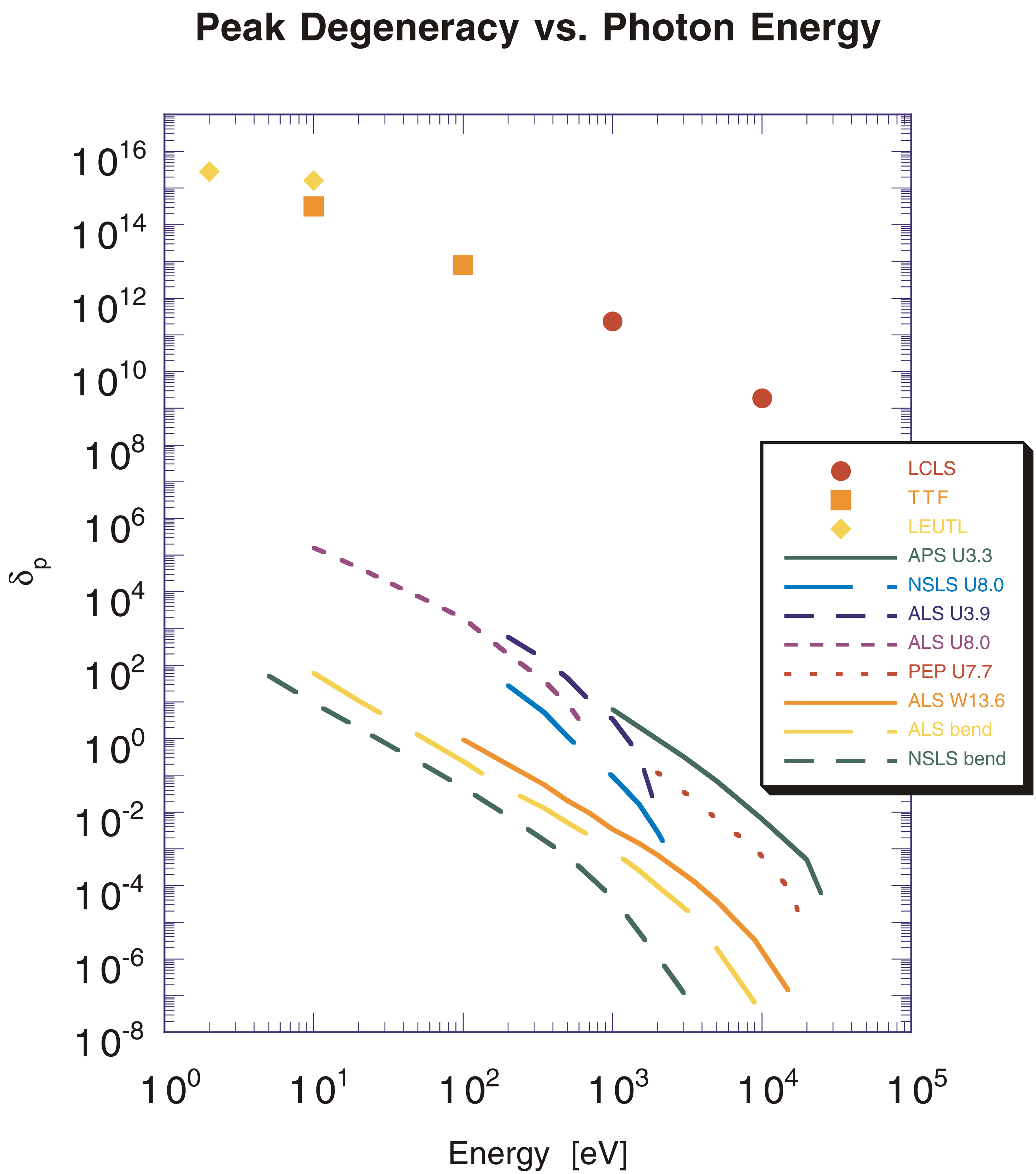
  Applications: structural biology, nanoscale materials science

- **Internal reference source**

  Use internal atom to form reference wave and object illumination

  Applications: atomic and molecular structure, soft condensed matter

	external reference	internal reference
reference source	zone plate	heavy atom
scattering type	elastic	fluorescence
intrinsic scattering	forward-directed	isotropic
resolution limit	optics, detector	λ, reference atom
feasible resolution	10-100 nm	10-100 pm
views needed for 3D	≥ 5	1
copies of structure	1	10^{10}
crystallinity required	no	no
structures oriented	-	yes
detector geometry	spherical	spherical
multiple energies	no	maybe



Peak Degeneracy vs. Photon Energy
10^{16}
10^{14}
10^{12}
10^{10}
10^{8}
10^{6}
10^{4}
10^{2}
10^{0}
10^{-2}
10^{-4}
10^{-6}
10^{-8}
δ_p
10^{0}
10^{1}
10^{2}
10^{3}
10^{4}
10^{5}
Energy [eV]
LCLS
TTF
LEUTL
APS U3.3
NSLS U8.0
ALS U3.9
ALS U8.0
PEP U7.7
ALS W13.6
ALS bend
NSLS bend

Flash sources

- **Realize greatest potential of x-ray holography**

- **Ideal requirements**

short pulses	< 1 ps to avoid radiation damage, blurring
high peak brilliance	> 10^{12} coherent photons in one pulse
high repetition rate	> 10 Hz
tunability	optimum sample contrast

- **Current and future sources**

	λ (nm)	t_{pulse} (ps)	B_{peak}
Rutherford XRL	23.6	200	10^{21}
LLNL XRL	20.6, 15.5, 4.5, 4.3	150	10^{22}
LCLS	*0.1 - 4*	*0.3*	10^{33}
Future FEL	*0.1 - 4*	*0.1*	10^{34}

Radiation dose and motion time scales

- **Absorbed dose processes scale as d^4 in 2D, d^6 in 3D**

process	dose (Gy)
kill hardy bacterium	10^4
chemical damage	10^5
ΔT = 100 K	4×10^5
vaporize water	2×10^6
vaporize everything!	10^7

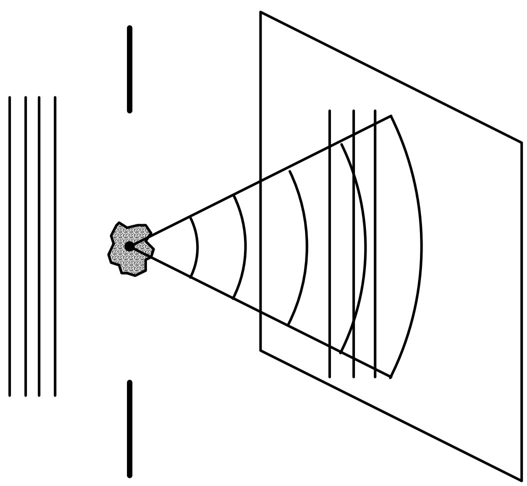
- **Several processes compete with imaging resolution, fidelity**

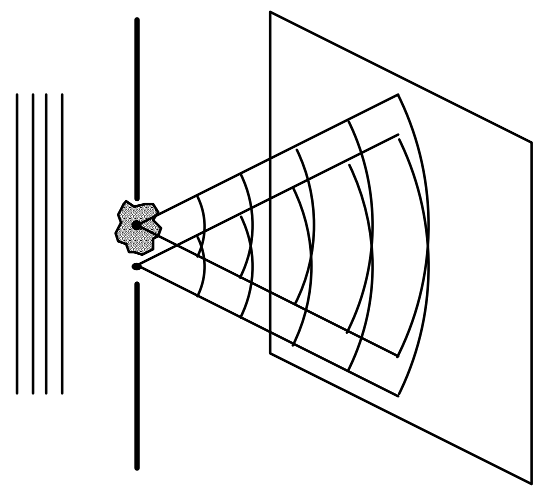
process	type	time scale (d~10 nm)
conductive cooling	exposure	10-100 ms
Brownian motion	natural	1-10 ms
biological dynamics	natural	~1 ms
chemical diffusion	exposure	10-100 μs
hydro. expansion	exposure	1-100 ps

- **Must obtain all structural information in one shot!**

J.C. Solem, *J. Opt. Soc.* **B3**, 1551 (1986)
R.A. London, et al., *SPIE Proc.* **1741**, 333 (1992)

X-ray holography: basic methods





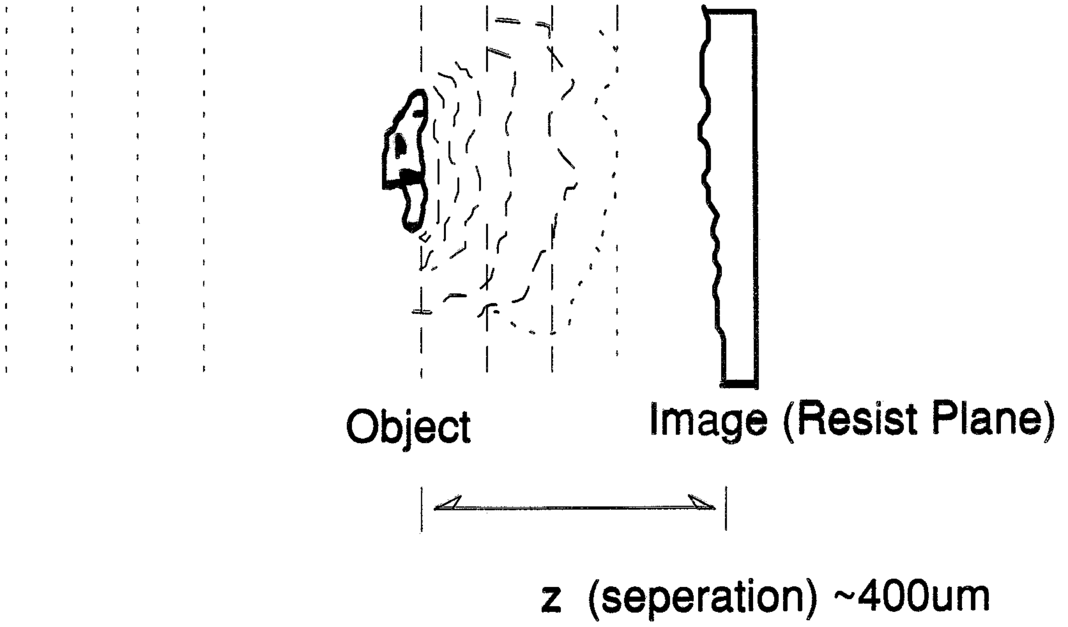
GABOR

plane reference
(in-line)

FOURIER TRANSFORM

spherical reference
(off-axis)

Gabor Holography



Advantages

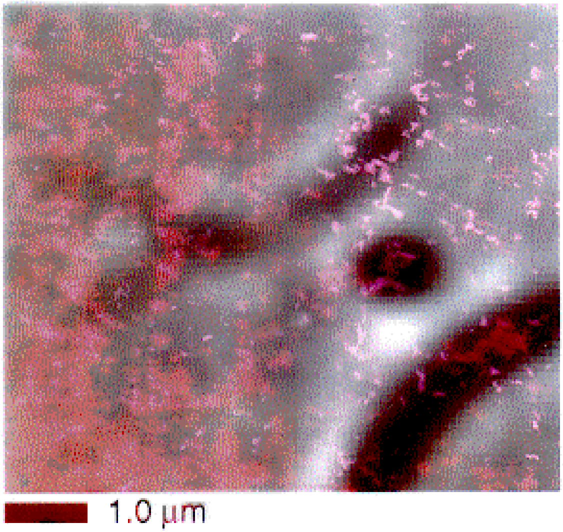
Pre-alignment (no focusing)
Separation not Critical
No Vibration Problem
Phase and Amplitude of Object
Insensitive to Beam Fluctuations
Ideal for Flash Sources

Disadvantages

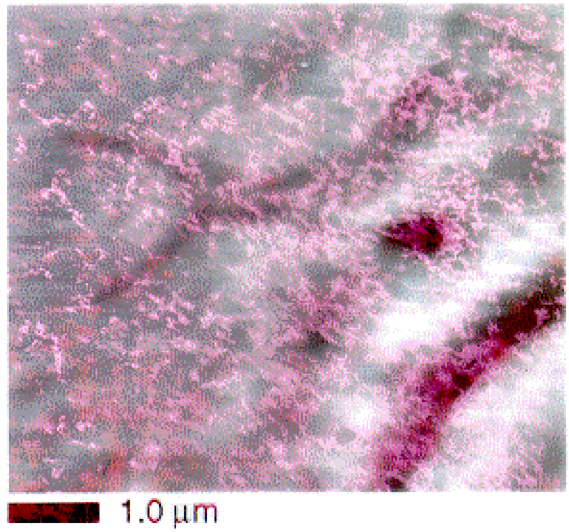
No magnification of Object
Long Exposure (with present sources)
Not Real-time
Twin-Image Noise

Gabor x-ray holography using photoresists

S. Lindaas and M. Howells, Advanced Light Source, LBNL
C. Jacobsen and A. Kalinovsky, SUNY at Stony Brook, Physics Dept.

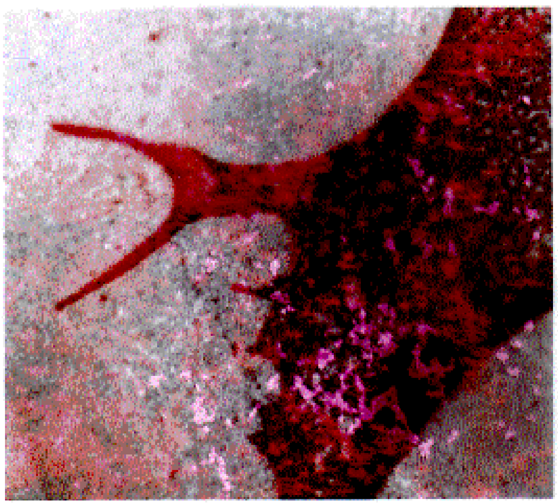


Visible light



X-ray

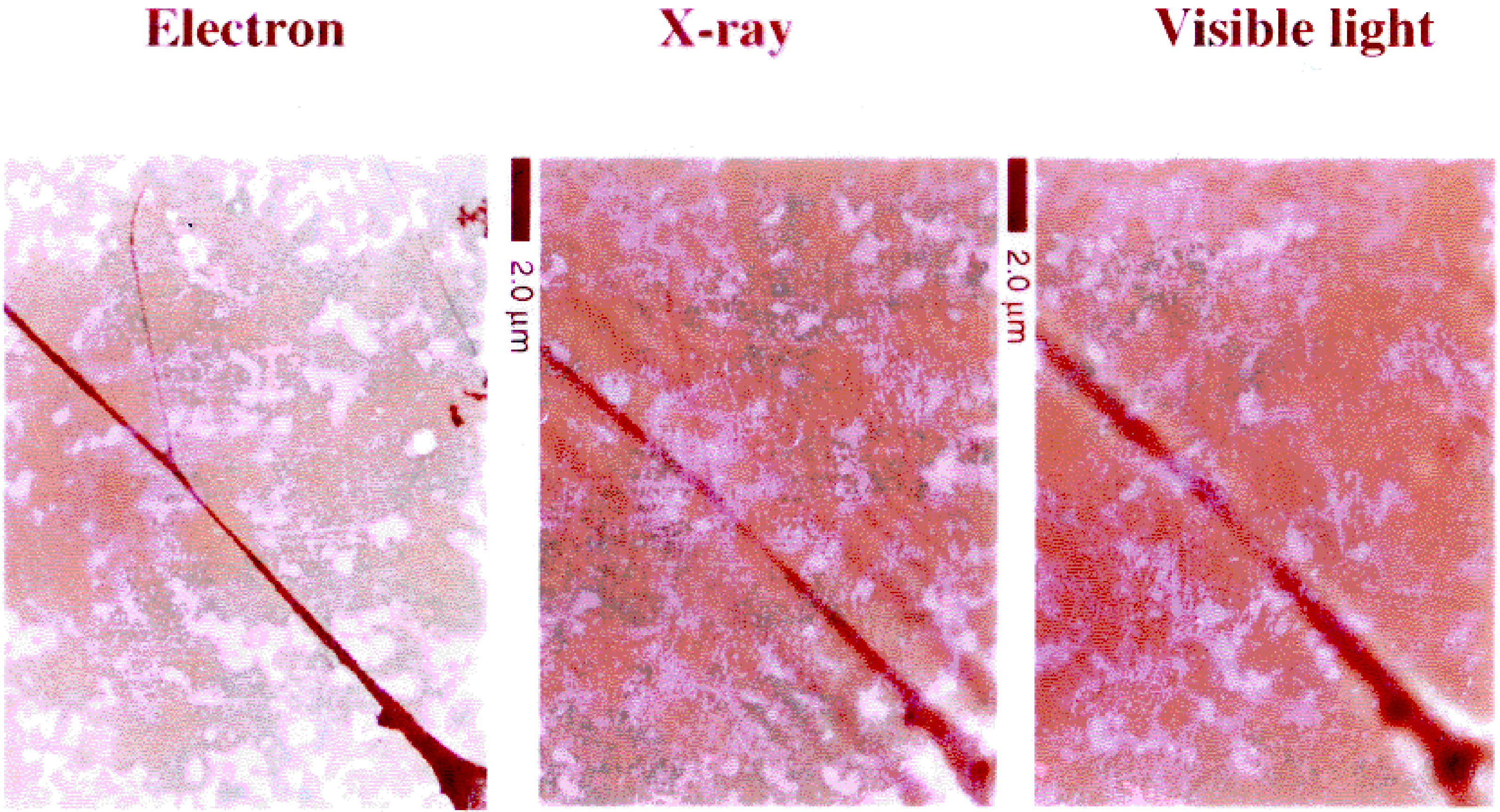
X-RAY HOLOGRAM
$\lambda = 1.89$ nm, $z = 425$ μm, readout using a custom linear-field scanning force microscope, numerical reconstruction.



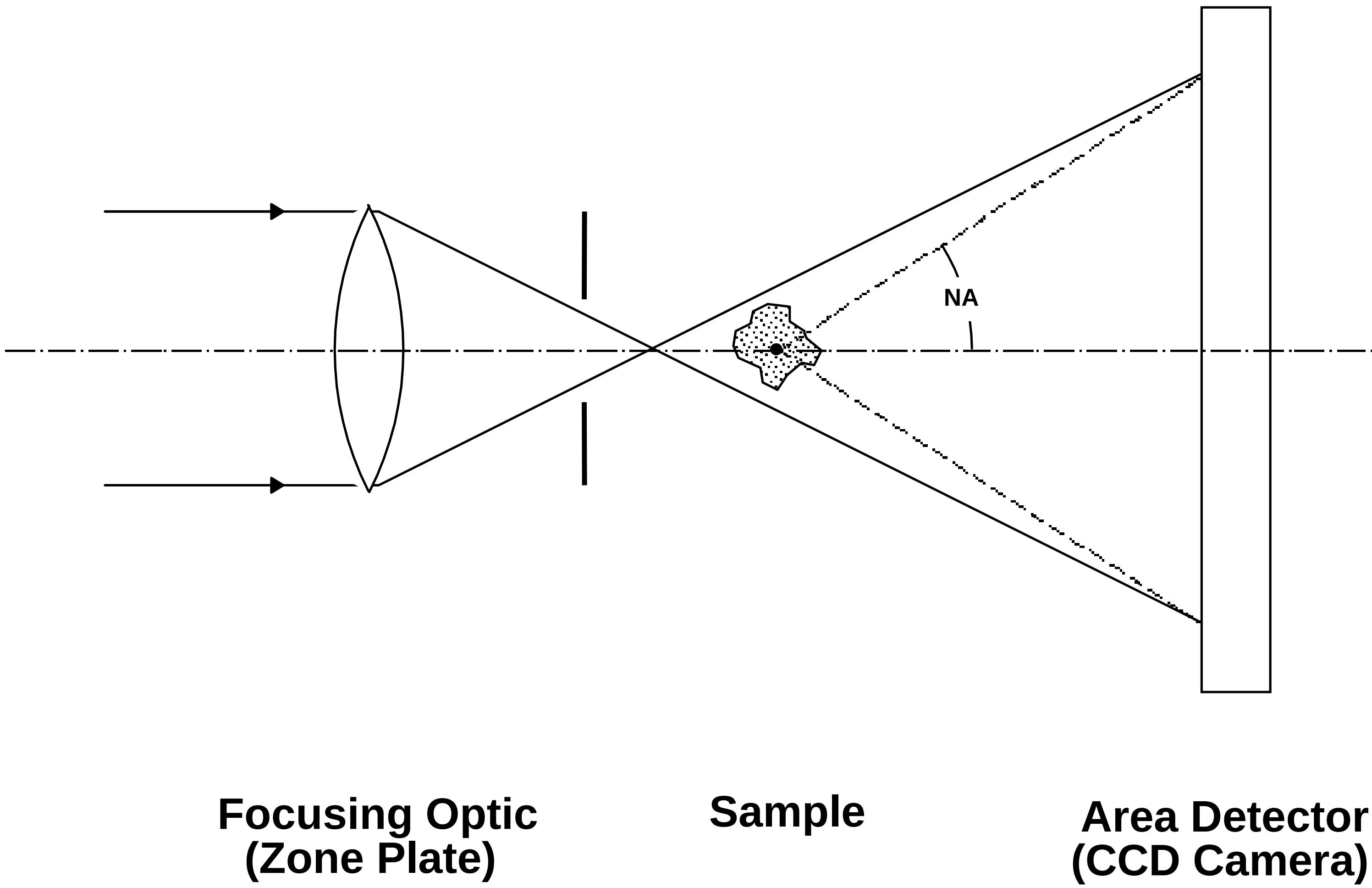
Electron

OPTICAL MICROGRAPH
100× Nikon objective, N.A.=0.90

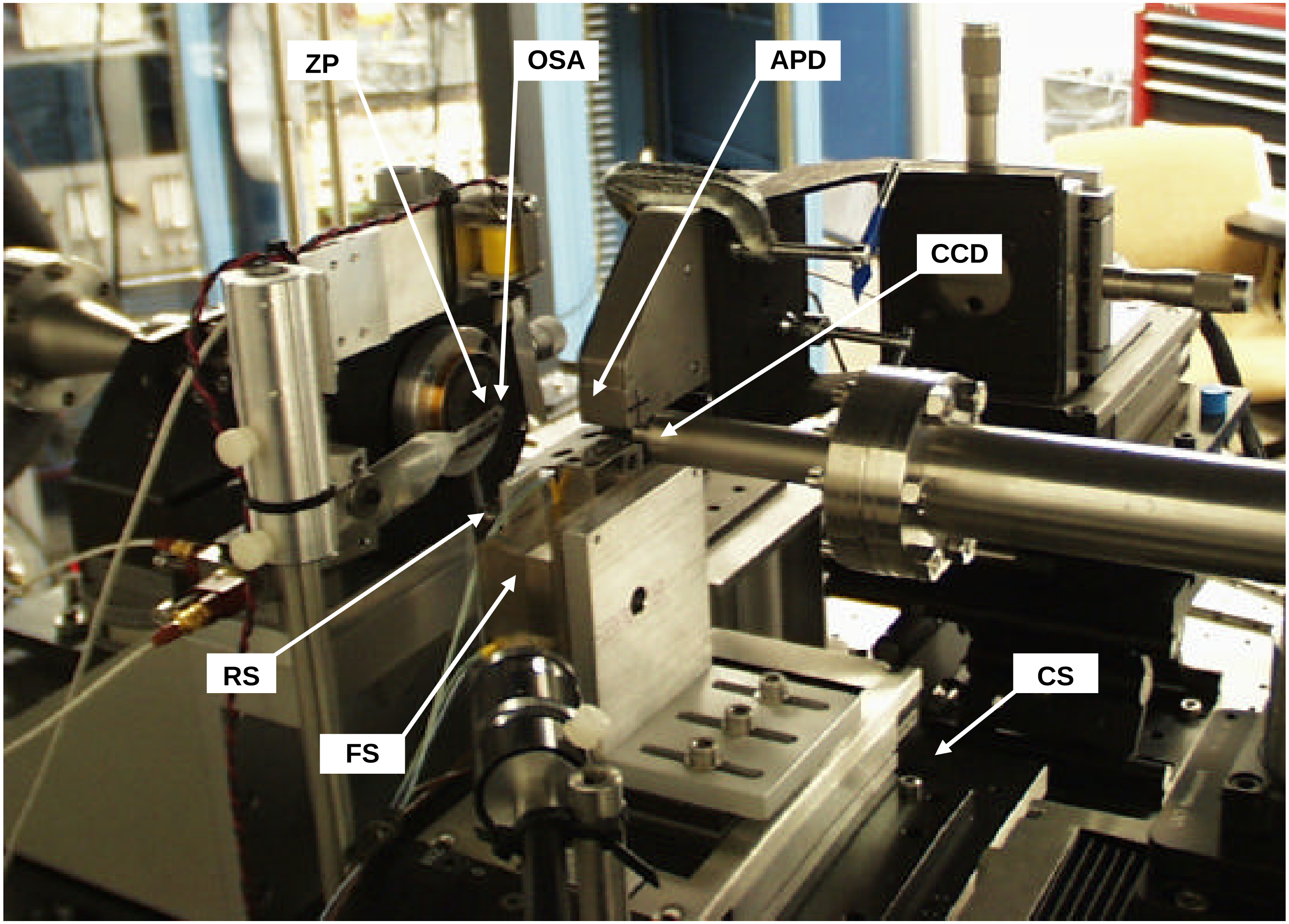
Images of a critical point dried NIL8 hamster neural fibroblast.


Electron
X-ray
Visible light
2.0 µm
2.0 µm

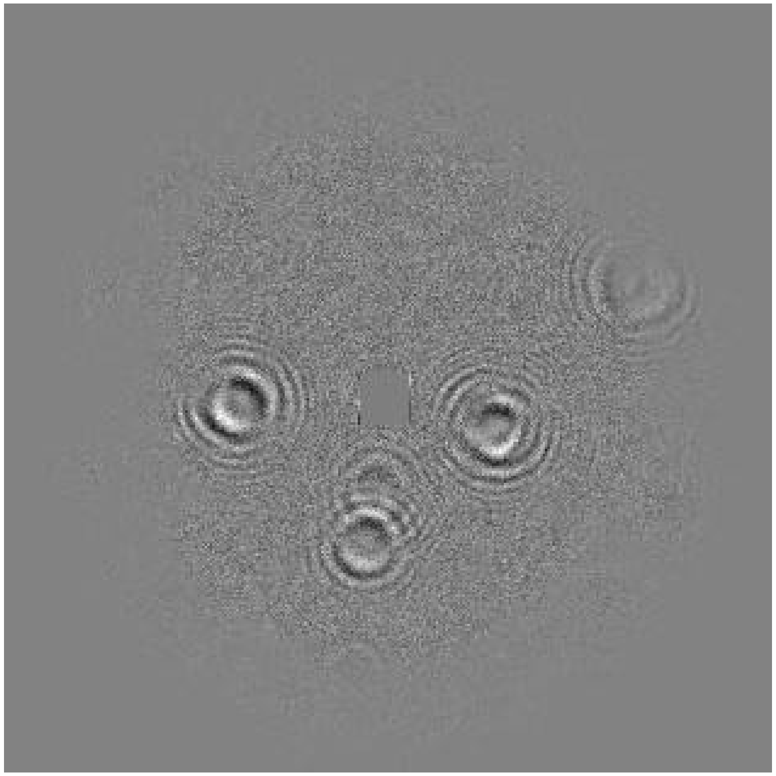
Fresnel transform holography



2-ID-B scanning/holographic microscope

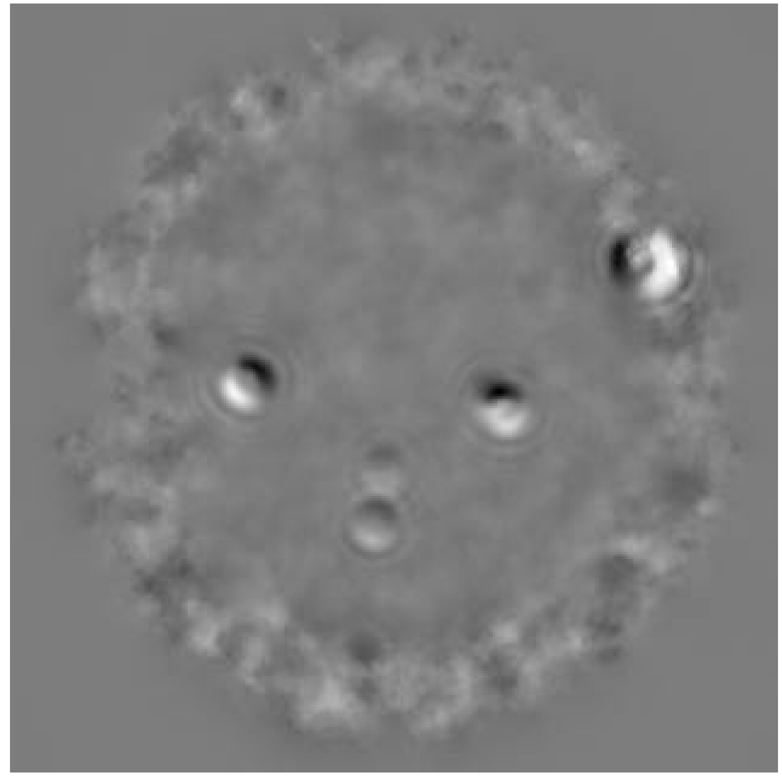


X-ray holographic phase imaging of 1-µm Al balls



5 µm

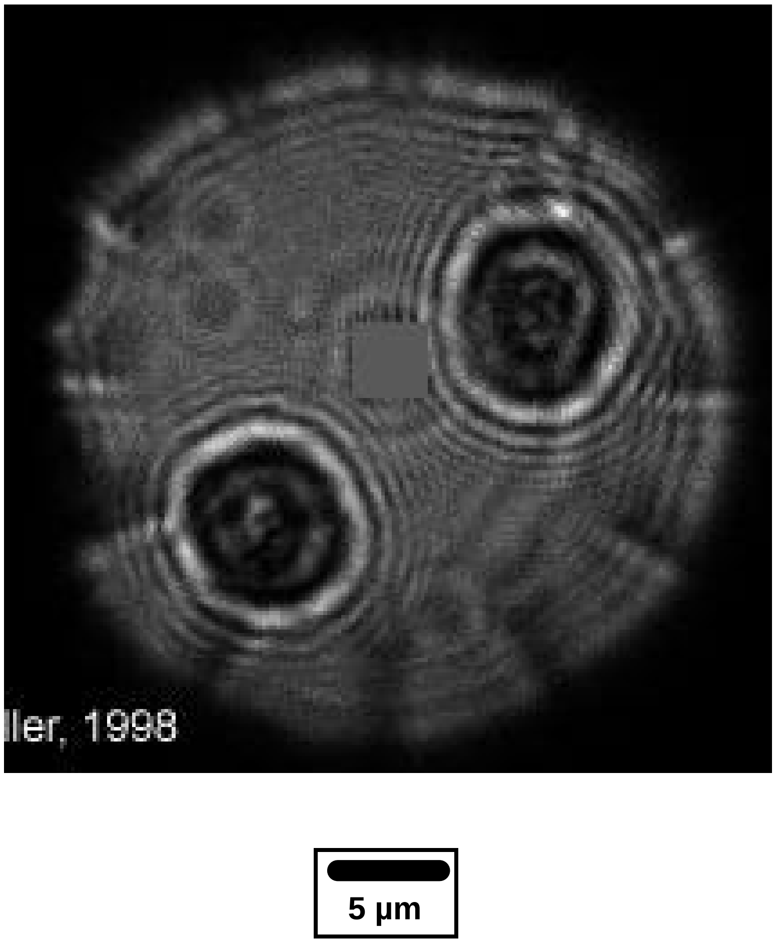
Normalized hologram



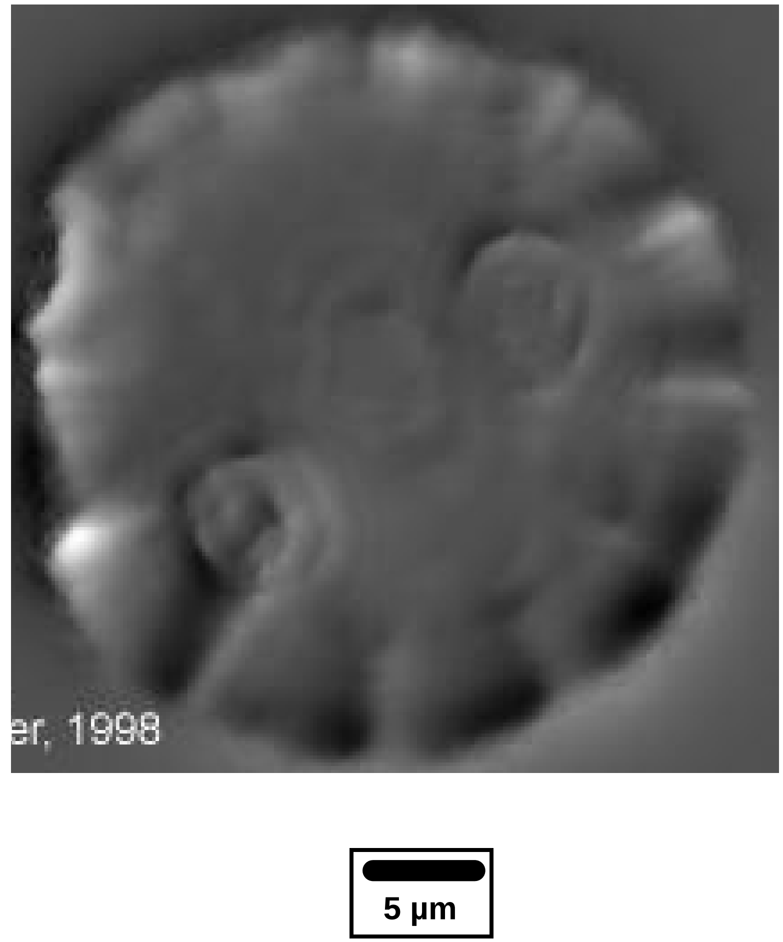
5 µm

Phase-reconstructed image

X-ray holographic phase imaging of ovarian cancer cells

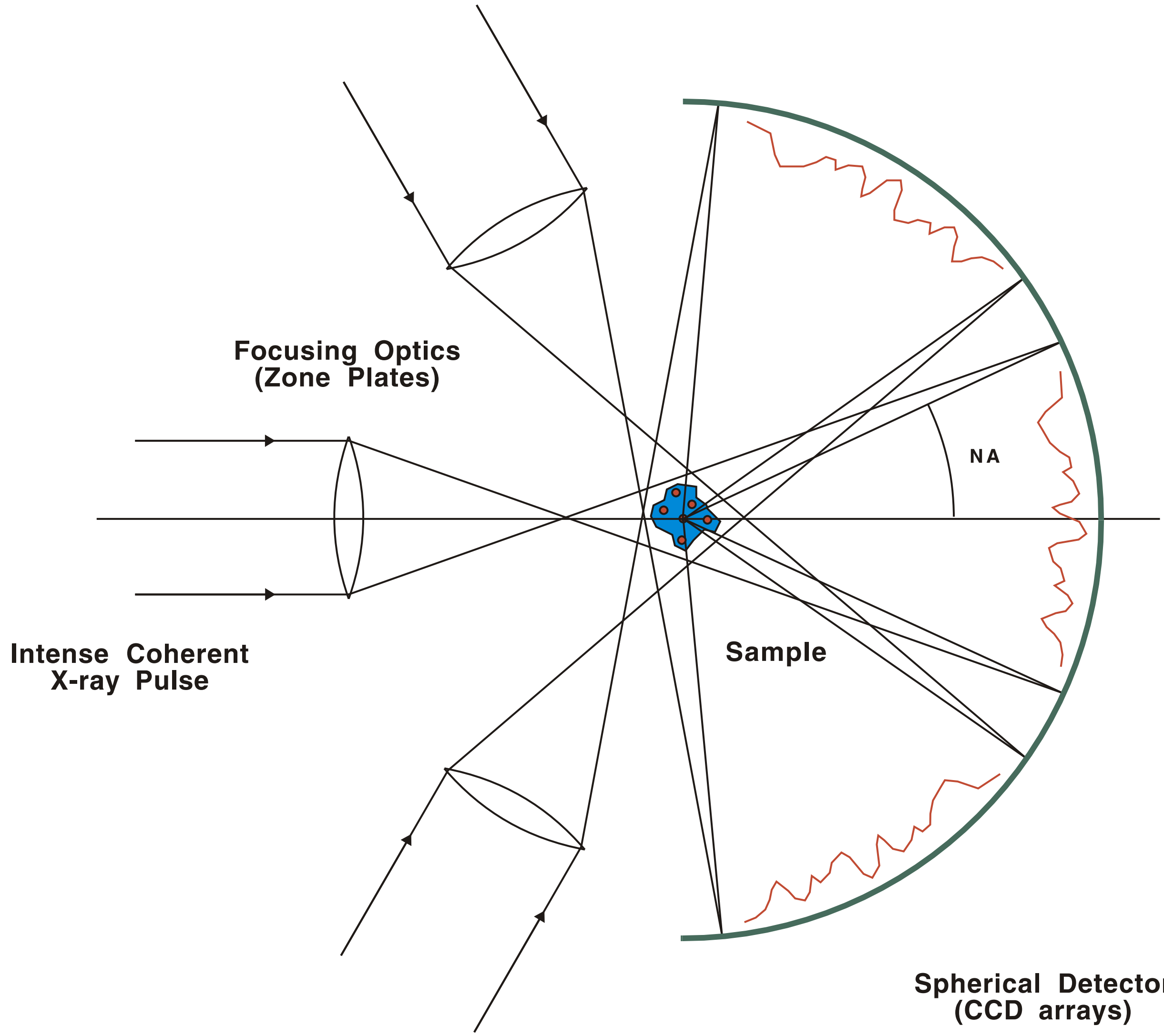


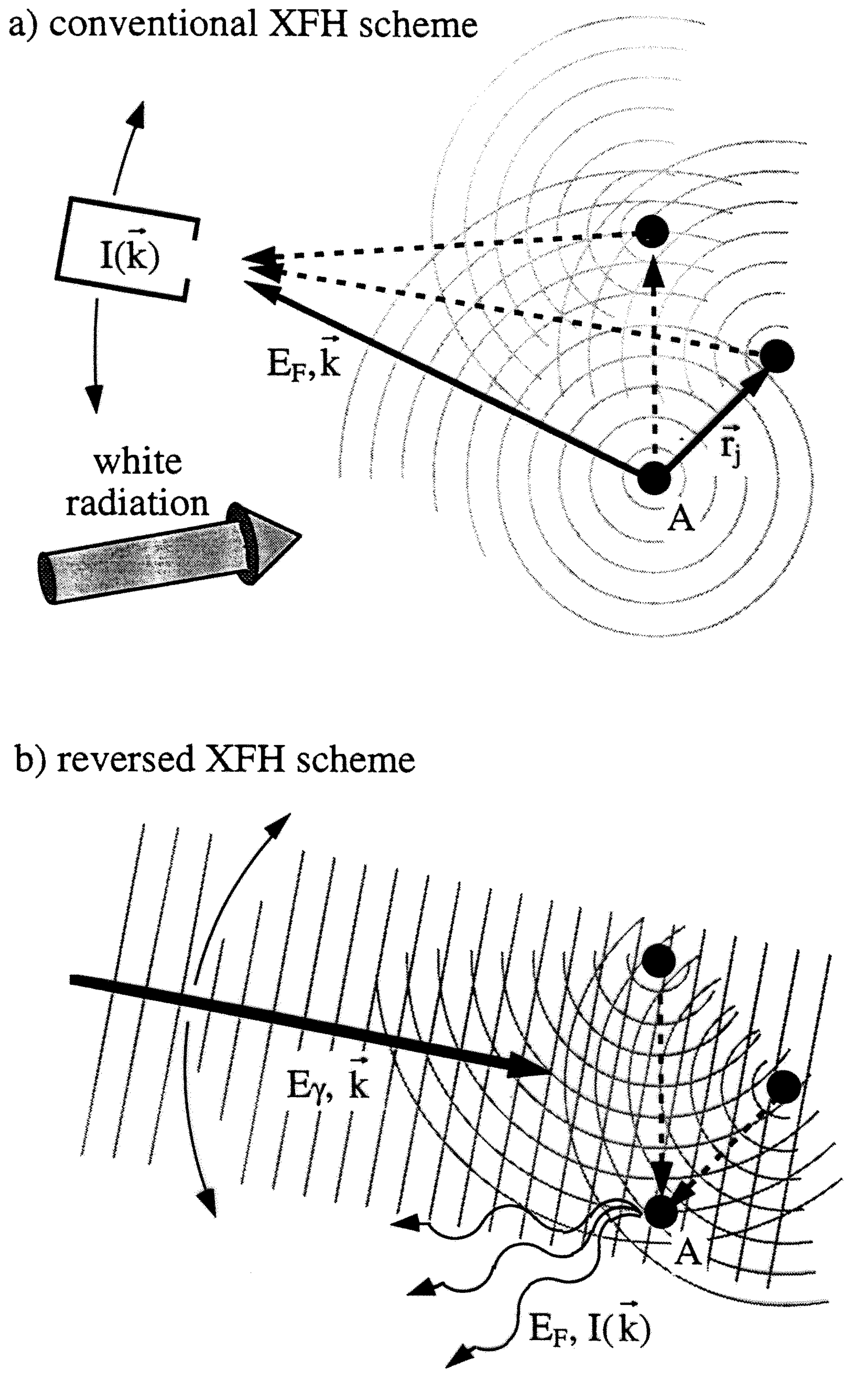
Normalized hologram



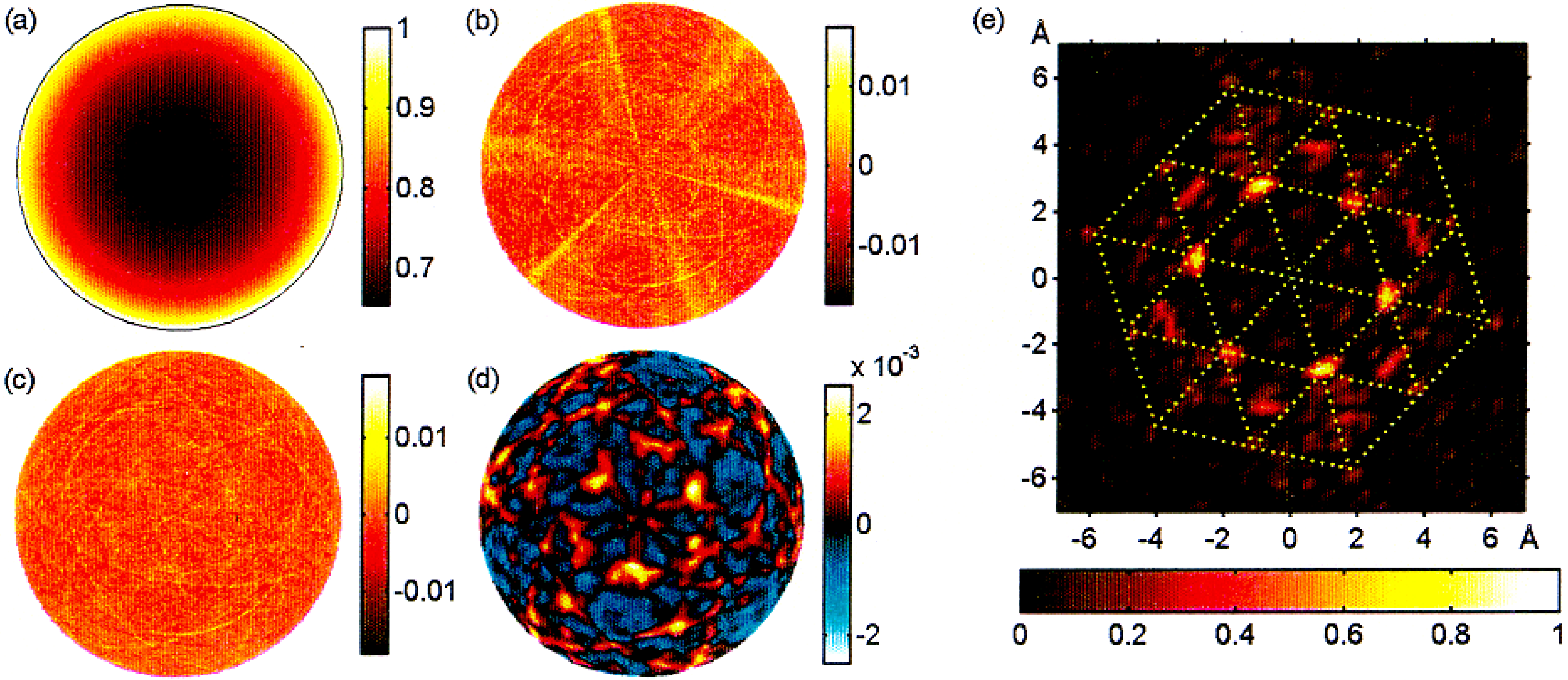
Phase-reconstructed image

Multiview Fresnel Holography



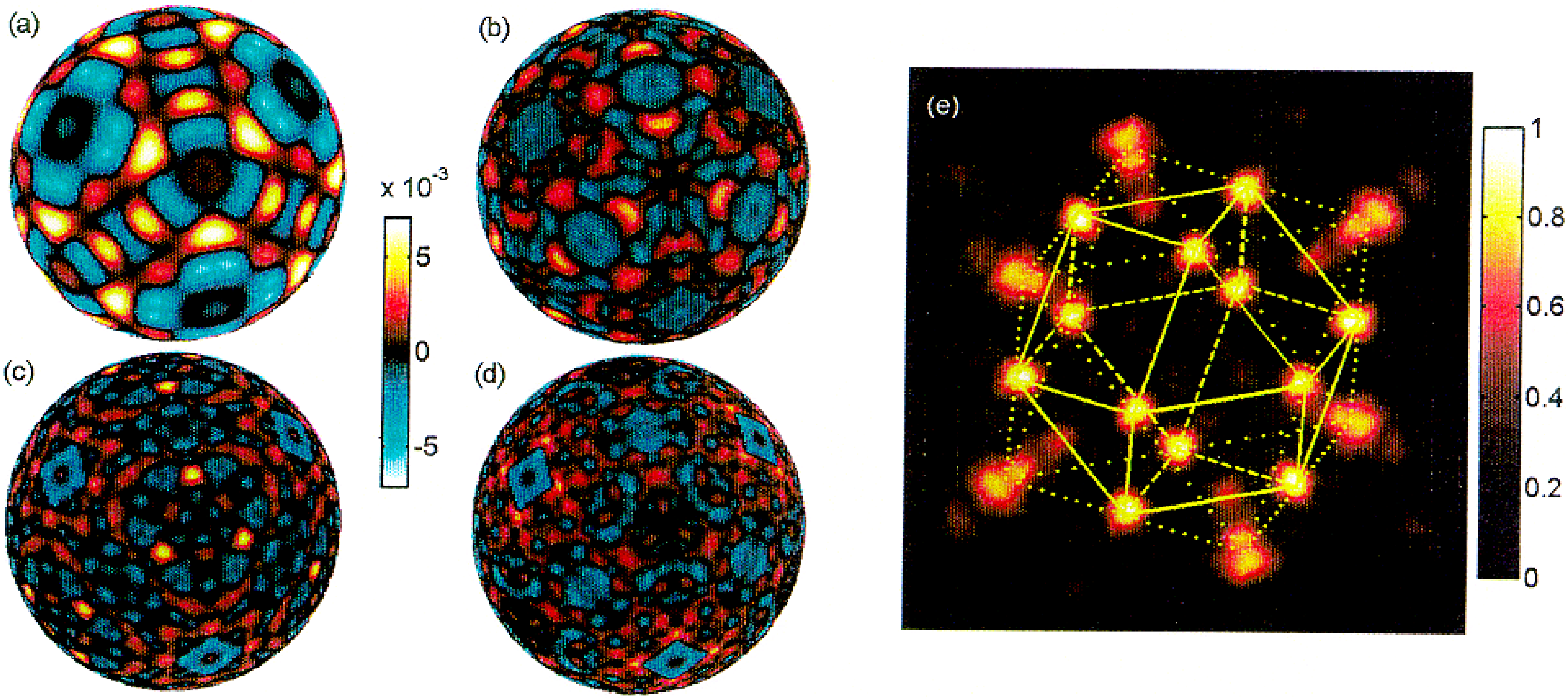


a) conventional XFH scheme
I($\vec{k}$)
$E_F, \vec{k}$
$\vec{r}_j$
white radiation
A
b) reversed XFH scheme
$E_\gamma, \vec{k}$
A
E_F, I($\vec{k}$)



(a)
1
0.9
0.8
0.7
(b)
0.01
0
-0.01
(c)
0.01
0
-0.01
(d)
x 10-3
2
0
-2
(e)
Å
6
4
2
0
-2
-4
-6
-6
-4
-2
0
2
4
6
Å
0
0.2
0.4
0.6
0.8
1

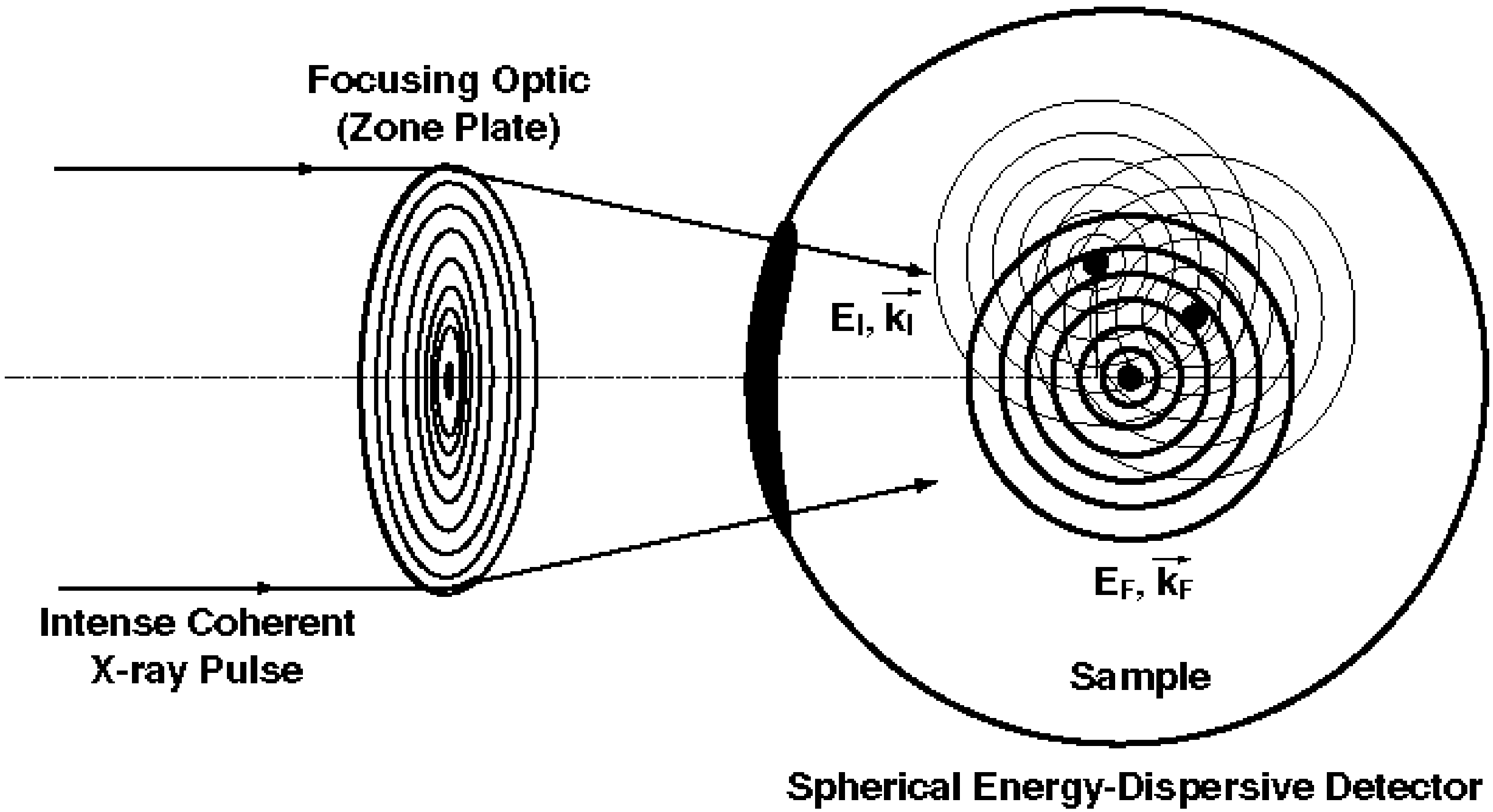
X-ray holography of CoO (fcc) at 50-pm resolution



holograms **reconstruction**

M. Tegze and G. Faigel, Europhys. Lett. **16**, 41 (1991)
T. Gog, et al., *Phys. Rev. Lett.* **76**, 3132 (1996)
M. Tegze, et al., submitted to Phys. Rev. Lett.

Atomic Resolution Holography



99.1.17

X-ray fluorescence holography: sample scaling

- XFH currently requires ~10^{20} copies of structure and ~10^{10} photons with third-generation source (B_{peak} ~ 10^{22})
- Expect ~10^{12} coherent photons/pulse with FEL (B_{peak} ~ 10^{33})
- Can focus FEL beam onto sample for intensity gain x 10^8

Have enough coherent flux in single FEL pulse to record 3D hologram of microscopic quantity of sample:

$$n_{structures} \sim 10^{20} / (10^2 \times 10^8) \sim 10^{10}$$

Challenges ahead

- **How to orient sufficient number of structure copies?**
  - intense pulsed laser fields
  - nanochannel arrays
- **How to build spherical, energy-resolving detector?**
  - polygonal CCD array
- **How to handle large, complex molecules?**
  - use heavy reference atom
  - multiple energies
  - over-determine problem
- **Can methods be extended to measure dynamics?**
  - use oriented, flowing media
  - synchronize experiment with pump laser
- **Can we utilize coherence of incident beam in XFH?**
  - use elastic scattering
  - stimulated fluorescence

Conclusion

- **X-ray holography is now feasible**
  - **50 nm resolution with external reference**
  - **50 pm resolution with internal reference**

- **Methods can be extrapolated to single-pulse regime**

- **Future x-ray sources will produce enough signal in one flash**

- **The potential and opportunities are exciting!**

Acknowledgements

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