Coherent X-ray imaging and dynamics of mezoscopic materials

Ivan Vartaniants

DESY, Hamburg, Germany National Research University, 'MEPhI', Moscow, Russia















High resolution microscopy



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High resolution electron microscopy



Simulated HREM images for GaN[0001]

From: Wikipedia



Atomic structure of the Au68 gold nanoparticle determined by electron microscopy.

M. Azubel, et al., Science 345, 909 (2014)



From: Web Images



Atomic resolution imaging of graphene

A. Robertson and J. Warner, *Nanoscale*, (2013),**5**, 4079-4093



From: Web Images



High resolution X-ray microscopy





Principles of X-ray microscopy

From: Web images







Figure 2 Images of a dandelion (taraxiacion officinale) collected with different analyzer settings (see text).





Can we develop x-ray microscope with atomic resolution?



It does not work with conventional approach due to the lack of atomic resolution X-ray optics

Can coherent x-ray diffraction imaging be the way to go?



Coherence



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What is Coherence?

Coherence is an ideal property of waves that enables stationary interference



Soap bubble



Whether X-rays are coherent waves?





Max von Laue Nobel Prize in Physics, 1914

W. Friedrich, P. Knipping, M. von Laue, Ann. Phys. (1913)

100 years later

LCLS







H. Chapman *et al.,* Nature (2011) A. Mancuso *et al.,* New J. Physics (2010)

Coherence



 $L_L = \lambda^2 / (2\Delta \lambda)$

 $L_T = (\lambda R / 2 D)$

Longitudinal Coherence Length

Transverse Coherence Length

Als-Nielsen & McMorrow (2001)

How to measure coherence?

I.A. Vartanyants & A. Singer "Coherence Properties of Third-Generation Synchrotron Sources and Free-Electron Lasers", Chapter in: Handbook on Synchrotron Radiation and Free-Electron Lasers

Young's double pinhole experiment



Hanbury Brown and Twiss experiment





Hanbury Brown & Twiss, Nature (1956)

Coherence properties of 3rd generation synchrotron sources

I.A. Vartanyants & A. Singer "Coherence Properties of Third-Generation Synchrotron Sources and Free-Electron Lasers", Chapter in: Handbook on Synchrotron Radiation and Free-Electron Lasers

Coherent volume (PETRA III, E=12 keV)

	L _V	$\mathbf{L}_{\mathbf{H}}$	L	Coh. Flux	
Raw Undulator	260 µm	40 µm	0.02 μm	6×10 ¹² ph/s	
We expect about two orders of magnitude more coherent flux at diffraction limited					h/s
Ra SOURCES					./ s
DC Si (111) High-β	280 µm	10 µm	0.5 μm	2×10 ¹⁰ ph/s	



Coherent X-ray Diffraction Imaging Lensless X-ray Microscopy



Coherent X-ray Diffraction Imaging in forward direction



Mancuso et al. J. Biotechnol. 149 229 (2010)

Small-angle scattering CDI Non-crystallographic samples **Uniform distribution of**

electron density

ASSOCIATION

Kinematical approximation:

$$A(q) = \int \rho(r)e^{-i\mathbf{q}\cdot r} dr$$

$$q_z = 0$$

$$A(q_x, q_y) = \int \langle \rho(x, y, z) \rangle_z e^{-i(q_x x + q_y y)} dx dy$$

$$\langle \rho(x, y) \rangle_z = \int \rho(x, y, z) dz$$

Inverse Fourier transform:

$$\langle \rho(x,y) \rangle_z = \frac{1}{(2\pi)^2} \int A(q_x,q_y) e^{i(q_x x + q_y y)} \,\mathrm{d}q_x \mathrm{d}q_y$$

Unfortunately, phases of $A(q_x, q_y)$ are not known!!!

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Phase retrieval



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Iterative phase retrieval algorithm



Real space constraints: use all *a priori* knowledge: •finite support •positivity **Reciprocal space constraint:** $|A_k(\mathbf{q})| \rightarrow \sqrt{I_{exp}(\mathbf{q})}$

R.W.Gerchberg & W.O. Saxton, *Optic* (1972) 35, 237
J.R. Fienup, *Appl Opt.* (1982) 21, 2758
V. Elser, *J. Opt. Soc. Am. A* (2003) 20, 40

Example of reconstruction (O.Yefanov)



Bragg Coherent X-ray Diffraction Imaging (Bragg CXDI)



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Coherent Scattering on Au Crystals



4 micron Au nanocrystal
Coherent X-ray Diffraction
Measurement at APS sector 33-ID (UNICAT)



I. Robinson, I. Vartanyants, et al., PRL (2001), <u>87</u>, 195505

Sensitivity to strain





Bragg CDI

Kinematical approximation:

$$A(\boldsymbol{q}) = \int \rho(\boldsymbol{r}) e^{-i\boldsymbol{q}\cdot\boldsymbol{r}} \,\mathrm{d}\boldsymbol{r}$$

$$\rho(\mathbf{r})$$
 – periodic electron density in crystal

Scattered amplitude near Bragg peak:

Shape function:

Phase, or projected strain field in crystal:

$$A_h(\boldsymbol{Q}) = \frac{F_h}{\nu} \int s(\boldsymbol{r}) \boldsymbol{e}^{-i\boldsymbol{Q}\cdot\boldsymbol{r}} \,\mathrm{d}\boldsymbol{r}$$

 F_h – structure factor

0 = a - h

$$\mathbf{s}(\mathbf{r}) = \begin{cases} 1, & \mathbf{r} \in \Omega \\ 0, & \mathbf{r} \notin \Omega \end{cases}$$

 $\varphi(\boldsymbol{r}) = \boldsymbol{h} \cdot \boldsymbol{u}(\boldsymbol{r})$

Crystallographic samples,

uniform distribution of strain

I.A. Vartanyants & I.K. Robinson, JPCM **13**, 10593 (2001); I.A. Vartanyants & O.M. Yefanov, in the book: *X-ray Diffraction Modern Experimental Techniques*. (2015), pp. 341-384.

3D Scan of the Reciprocal Space





 $q_z = -2.8 \times 10^{-3} \text{ nm}^{-1}$

 $q_z=0$

 $q_z = +2.8 \times 10^{-3} \text{ nm}^{-1}$

G. Williams et al., PRL (2003) 90, 175501

3D mapping of a deformation field inside a nanocrystal





Diffraction patterns from Pb nanocrystal measured around (111) Bragg peak

Strain field shown by color gradient Resolution: 40÷50 nm

M. Pfeifer, *et al.*, Nature, <u>442</u>, 63 (2006), R. Harder, *et al.*, PRB B **76**, 115425 (2007).

Coherent Diffractive Imaging



of the sample

iants | Condensed Ma

Crystallographic samples **Uniform strain field** distribution in the sample

Coherent imaging with atomic resolution



 J. Gulden, et al.,
 "Imaging of Nanocrystals with Atomic Resolution Using High-Energy Coherent Xrays", XRM-2010 Proceedings, AIP Conf. Proc. 1365, 42-45 (2011)

Theory

If several Bragg peaks are measured simultaneously:

$$A(\boldsymbol{q}) = \int \rho(\boldsymbol{r}) e^{-i\boldsymbol{q}\cdot\boldsymbol{r}} \, \mathrm{d}\boldsymbol{r} = B(\boldsymbol{q}) \cdot F(\boldsymbol{q}) \cdot [\rho_{\infty}(\boldsymbol{q}) \otimes s(\boldsymbol{q})]$$

Here: B(q) – envelope function; F(q) – structure factor of a unit cell; s(q) – FT of the shape function

$$\rho_{\infty}(\boldsymbol{q}) = \frac{(2\pi)^3}{\nu} \sum_{\boldsymbol{h}} \delta(\boldsymbol{q} - \boldsymbol{h})$$

Inverse Fourier transform of this relationship:

 $\rho(\mathbf{r}) = [b(\mathbf{r}) \otimes \rho_{uc}(\mathbf{r})] \otimes [\rho_{\infty}(\mathbf{r}) \cdot s(\mathbf{r})]$

Here: $b(\mathbf{r}) - FT$ of the envelope function $B(\mathbf{q})$; $\rho_{uc}(\mathbf{r})$ – electron density of a unit cell; $s(\mathbf{r})$ – shape function

$$\rho_{\infty}(\mathbf{r}) = \sum_{n} \delta(\mathbf{r} - \mathbf{R}_{n});$$
$$\mathbf{R}_{n} = n_{1}\mathbf{a}_{1} + n_{2}\mathbf{a}_{2} + n_{3}\mathbf{a}_{3}$$

I.A. Vartanyants & O.M. Yefanov, in the book: *X-ray Diffraction Modern Experimental Techniques*. (2015), pp. 341-384.

Theory

This electron density is peaking at the regular positions of the unit cell due to the function $\rho_{\infty}(\mathbf{r})$ and has an overall shape of the sample $s(\mathbf{r})$.

Most importantly, it contains the position of the atoms in the unit cell due to the reconstruction of the electron density function of a unit cell $\rho_{uc}(\mathbf{r})$.

This means the following: If the continuous intensity distribution around several Bragg peaks will be measured simultaneously and phase retrieval methods will be applied to get the phase, then, in principle, the electron density with *atomic* resolution will be obtained.

I.A. Vartanyants & O.M. Yefanov, in the book: *X-ray Diffraction Modern Experimental Techniques*. (2015), pp. 341-384.

CXDI using high energy coherent X-rays



J. Gulden, et al.,

"Imaging of Nanocrystals with Atomic Resolution Using High-Energy Coherent X-rays", *XRM-2010 Proceedings*, AIP Conf. Proc. **1365**, 42-45 (2011)

CXDI using high energy coherent X-rays





F=10¹³ photons

(111) (000)10 10¹³ photons (c)

5 nm

(111) (000)10 10¹⁴ photons 10-1 (b)

10

F=10¹⁴ photons

5 nm (f)

F=10¹⁵ photons







(h)

5 nm

Pd nanocrystal 10 nm size

(g)



An excellent scientific goal for diffraction limited storage ring?


First realization of these ideas with photonic (colloidal) crystals





Photonic (colloidal) crystals





Photonic crystals

Photonic crystals in nature





Artificial photonic crystals



Photonic band gap materials





lambda [nm] https://www.theochem.kth.se/research/phot (Condensed Watter School, Zelenogorsk, Warch 16, 2016 | Page 39 t/Photonic_Crystals.html

Photonic crystals (defects)

- Semiconductors in the 20th century, now photonic century?
- Photonic crystals- optical materials for the 21st century
- Artificial defects can be used as wave guides



Simulation of a lightwave in a photonic crystal composed of rods

Jasmin Smajic et al., Optics Express **11**, 1378 (2003). SEM image of the crystal surface of a colloidal crystal in the presence of the stacking fault.

SEM images of the internal facets of silicone inverse opal. **a** [110] facet **b** [111] facet

Gulden et al., Phys. Rev. B (2010) 81 224105

Alvaro Blanco et al., Nature 405, 437 (2000).

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Colloidal crystals grown by self-organization



Growth of the colloidal crystal film

Vertical deposition method







J-M. Meijer, et. al. Langmuir (2012), 28, 7631–7638.

Polystyrene colloidal crystal



Coherent x-ray imaging of defects in colloidal crystals

J. Gulden, et al., Phys. Rev. B 81, 224105 (2010)

Measurements with coherent x-rays at 3rd generation synchrotron sources







European Synchrotron Radiation Facility ESRF

Measurements at azimuthal angle $\varphi = 0^{\circ}$



Experimental conditions (ID06, ESRF):

- Energy: E=14 keV
- Pinhole size 6.9 µm
- Sample detector distance: 3.96 m
- Detector pixel size: 9 µm

ELMHOLTZ • Detector size: 4005 × 2671 pixels

J. Gulden, et al., Phys. Rev. B 81, 224105 (2010)

Measurements at azimuthal angle $\varphi = 0^{\circ}$



Diffraction pattern with the subtracted pinhole

ELMHOLTZ ASSOCIATION

Results of reconstruction from this diffraction pattern





(c)

Measurements at azimuthal angle φ = 35°



Experimental conditions (ID06, ESRF):

- Energy: E=14 keV
- Pinhole size 6.9 µm

HELMHOLTZ

- Sample detector distance: 3.96 m
- Detector pixel size: 9 μm
- **ASSOCIATION** Detector size: 4005 × 2671 pixels



Measurements at azimuthal angle $\varphi = 35^{\circ}$





For the first time defect core was directly imaged using coherent x-rays

Extension to 3D

Crystallography with coherent X-rays

J. Gulden, et al., Optics Express, 20(4) 4039 (2012)
A. Shabalin, et al., (2015) (in preparation)



PETRA III





Structural evolution of colloidal crystal films in the vicinity of the melting transition



Tuning of properties by external fields

Pumping energy

Strain field engineering





Y. Y. Hui et al., ACS Nano 7 (8), 7126 (2013)



Incremental heating



M. Daryl et al., PRL 108, 033902 (2012)

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http://www.st-andrews.ac.uk/microphotonics/Gallerv.php

http://www.amolf.nl/research/nanooptics/news/detailpage/artikel/pumped-photonic-crystal-accelerates-slow-light/



Schematics of the In Situ High-Resolution X-ray Scattering Setup at the P10 Coherence Beamline of the PETRA III Light Source (see p. 5A)

ACS Publications Most Trusted. Most Cited. Most Read.

E. Sulyanova et al., Langmuir **31**(19), 5274 (2015).

www.acs.org

Experimental setup. P10 beamline, PETRA III



X-ray diffraction patterns measured *in situ* during incremental heating

Experiment A

15 keV, 50 x 50 μm unfocused beam





Experiment B 8 keV, 3.5 x 2.8 μm focused coherent beam



X-ray diffraction pattern of the experiment A measured at room temperature



X-ray diffraction pattern of the experiment A measured at room temperature. Same pattern with SAXS contribution subtracted. Enlarged area of (b) showing Bragg peak indexing.



Temperature evolution of the Bragg peaks parameters experiment A



Т, К

Integrated intensity



Temperature evolution of the Bragg peaks parameters experiment A



Williamson-Hall plot



Williamson-Hall plot





Williamson-Hall plot



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Temperature dependence of SAXS curves



Experiment A

15 keV, 50 x 50 mm unfocused beam Experiment B 8 keV, 3.5 x 2.8 μm focused beam



Nano- scale

Mesoscopic scale



Polysterene particle diameter *D* and average lattice parameter

 $< a_{[110]} >$

Lattice distortion parameter g_q and domain misorientation parameter g_{φ}



The model of colloidal crystal melting process

Nano- scale





Mesoscopic scale



The model of colloidal crystal melting process

Nano- scale



Mesoscopic scale



Free-electron lasers



We are living in exciting time !

FLASH at DESY 2005

LCLS at Stanford 2009





Sometime Nobel prize in physics?



2011

European XFEL under construction (2016-2017)

European XFEL







Financial participation (2005 Euro)






Pump-probe experiments at FELs





Study of dynamics in colloidal crystals



Observation and tuning of hypersonic bandgaps in colloidal crystals

- > Polysterene spheres in air, glycerol, PDMS and silicon oil
- > D = 256 nm, 307 nm
- > Brillouin spectroscopy
- > No sintering



Supported opal and scattering geometry



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Hypersonic modulation of light in three-dimensional photonic and phononic band-gap materials

- > Silica spheres, 359 nm diameter, 10-12 layers
- > IR energy converted into vibrations with an 100 nm thick AI foil ("hypersonic transducer")
- > Sintered crystal, coupling parameter $\chi = D/2a 1 = 0.015 \pm 0.005$
- > Reflectivity measurements





Reflectivity measurements

Akimov et al., Phys. Rev. Lett., (2008)

Set up for pump-probe experiment

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Pump-probe experiment on colloidal crystals at FLASH





Pump probe experiment on colloidal crystal film at FLASH

> Study of colloidal crystal in the temporal domain

- ✓ Elastic vibration of the spheres (Lamb modes)
- ✓ Collective vibrations (phonons)
- ✓ Order-disorder transitions

Pump-probe experiment:

- > Pump: 800 nm IR laser
- > Probe: 8 nm FEL radiation
- > Time delay from -100 ps up to 1000 ps, with 50 ps steps



R.Dronyak et. al., Phys Rev B 86 (2012)

Pump-Probe Experiment on Colloidal Crystal Film at FLASH



Single-shot diffraction patterns at different time delay

↓10⁴ = 10³ 220 -10² 1/3(422) -10^{1} 2/3(422 20 µm 10^{0}

The momentum transfer vector \mathbf{Q} and the horizontal W_x and vertical W_y size of the peaks were analyzed

R.Dronyak et. al., Phys Rev B 86 (2012)

Pump-Probe Experiment on Colloidal Crystal Film at FLASH



selected 2/3(422) and 220 Bragg peaks

Theoretical calculations of vibrations of a 400 nm isotropic elastic sphere based on the Lamb theory reveal a 5.07 GHz eigenfrequency of the ground (breathing) mode



Pump-probe experiment on colloidal crystals at LCLS

1njector/Linat 600m e accelerator (SLAC) er Beam Transport 227m above ground facility to transport electron beam (SLAC)

- Undulator Hall 170m tunnel housing undulators (ANL)

Electron Beam Dump 40m facility to separate e and x-ray beams (SLAC)

Front End Enclosure 40m facility for photon beam diagnostics (LLNL)

Near Experimental Hall: 3 experimental hutches, prep areas, and shops (SLAC/LLNL)

X-Ray Transport & Diagnostic Tunnel 210m tunnel to transport photon beams (LLNL)

Far Experimental Hall 46 cavern with 3 experimental hutches and prep areas (SLAC/LLNL)

Pump-Probe experiment on colloidal crystals

Experimental setup



Experimental setup@XPP



Experimental setup@XPP



CSPAD detector



Experimental setup@XPP



Parameters of X-ray and IR laser beams

1. X-ray beam

- E=8 keV
- Pulse duration: \leq 50 fs
- Flux_{sample} ~10⁹ ph/pulse
- Focus ~ 50 μm
- Energy bandwidth ~10⁻⁴

2. Laser beam

- $\lambda_{las} = 800 \text{ nm}$
- Pulse duration: \leq 50 fs
- E ~ 2 mJ
- Power: P ~ 4*10¹⁰ W
- Focus ~ 100 μm





Pump-Probe experiment on colloidal crystals



Pump-Probe on Colloidal Crystals

	Camera: XPP Gige 6		
Cameras Show/Hide Data Processing Orientation Zoom Markers/ROI Administration			Camera Camera XPP Gige 6 Connected YES Data Rate 3.4 Hz Display Rate 3.4 Hz Color Map
			Hot Log Scale Min Max Force Color Image to Grayscale Display Single Frame (at -5 Hz)
	•	•	○ Local Average (at 5Hz / #) 1 Marker 1 × 454) Y 497 2 × 1334) Y 863 3 × 1375) Y 70
	0		4 X 916 Y 483 Region of Interest X 0 Y 0 W 1388 H 1038 [Set ROI] Reset ROI Zoom Zo
	•		Zoom In (2x) Zoom Out (0.5x) Zoom To ROI Zoom to Actual Size GigE Camera Settings Camera Mode Fixed Rate Gain 1 Acquisition Time (s) 0.3
			Acquisition Period (s) 0.2

an 240.63 Std 98.28 Var/Mean 40.14 (0,0) W 1388 H 1038 t# 1/1 Color scale [0,741] Zoom 3.138 7): 143 1:(454,497): 345 2:(1334,863): 130 3:(1375,70): 239 4:(916,483): 666

Damage produced by laser power 7° wave plate angle (~50 µJ)

Study of ultrafast melting of colloidal crystals



Decay of integrated intensity of the peaks

Diffraction pattern



Decay of integrated intensity



τ - time delay between laser and X-ray pulses

$$\frac{\Delta I(\tau)}{\langle I \rangle} = A \cdot exp\left(-\frac{\tau}{\tau_0}\right) - A$$

Fit of integrated intensity decay with exponential function



Time delays from -10 ps to +1000 ps $\Delta \tau$ =25.25 ps

Time delays from -10 ps to +250 ps $\Delta \tau = 6.5$ ps



Bragg peak's broadening in q-direction



Bragg peak's broadening in q-direction



1. Energy transfer from IR laser to colloidal crystal?

2. Response of colloidal crystal lattice?

- a. Decay of Bragg peaks integrated intensity
- b. Growth of Bragg peaks FWHM



Model of ultrafast melting of colloidal crystal

Energy transfer from IR laser to colloidal crystal

- IR wavelength: 800 nm
- Energy: 1.5 eV
- Energy of chemical bonds: (C-C, C-H) ~3-4 eV
- Absorption coefficient of 800 nm radiation in polystyrene: 10⁻⁴
- Temperature raise: one-two degrees



Response of colloidal crystal lattice

Colloidal crystal is heated above T_g in 50 fs



Soft PS spheres



Response of colloidal crystal lattice

Colloidal crystal is heated above T_g in 50 fs In ≤50 ps a sintering of PS spheres is going



Soft PS spheres

Sintering of PS spheres



Response of colloidal crystal lattice

Colloidal crystal is heated above T_g in 50 fs In ≤50 ps a sintering of PS spheres is going In few 100 ps PS dynamics is damped (due to viscosity)



Soft PS spheres Sintering ofLiquid to solidPS spheresIIIIIItransition

age 100 DESY

Model of ultrafast melting of colloidal crystal

This model explains decay of integrated intensity but does not explain the broadening of the Bragg peaks



The broadening of Bragg peaks can be explained by including the imperfections of the colloidal crystal lattice in the model





Model of ultrafast melting of colloidal crystal

Work in progress ...



DESY future projects for large scale facilities

FLASH2020



further experimental hall
8 experiments

......

Courtesy to E. Weckert

PETRA IV: scope / motivation / design



ivan varianianis | Condensed Watter School, Zelehogorsk, Warch To, 20.2, 20.2 Jenu

PETRA IV roadmap (long term)

- Covering tender to very hard X-rays
 5 7 GeV electron energy
- Increase the coherent fraction by at least one order of magnitude
 aim for ~10-20 pmrad emittance (extrapolation of present day techniques)
- Increase number of undulator ports to ~30
 - ➔ additional experimental hall



Time frame: from 2024/2025 on



PETRA IV: status and further development



Coherent X-ray Scattering and Imaging Group at DESY

Present members:

•	S.	Lazarev
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- I. Besedin
- P. Skopintsev
- D. Dzhigaev

- I. Zaluzhnyy
- M. Rose
- O. Gorobtsov
- N. Mukharamova



Former members:

- A. Zozulya (now@PETRA III)
- A. Mancuso (now@XFEL)
- O. Yefanov (now@CFEL)
- R. Dronyak
- J. Gulden (now@FH-Stralsund)
- U. Lorenz (now@University of Potsdam)
- A. Singer (now@UCSD)
- R. Kurta (now@XFEL)
- A. Shabalin (now@CFEL)

Acknowledgements

- **DESY**
 - E. Weckert
 - Former and present members of my group
 - P10 beamline (M. Sprung, A. Zozulya)
 - FLASH team
- University of Utrecht
 - A. Petukhov
 - J.-M. Meijer
- LCLS
 - **XPP** beamline
- Russia
 - Institute of Crystallography RAS
 - RC "Kurchatov Institute"
 - National Research Nuclear Centre, "MEPhI"

We are looking for motivated PhD students and PostDocs

Thank you for your attention!



