H2020-MSCA ITN
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Stockholm University

Electron Pair Distribution Function (ESR7)

Cheuk-Wai Tai – Stockholm University

NanED| Joint Initial Meeting   Pontedera, 29st- 30st November 2021

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ESR	Supervisory Team	
	Main Supervisor	Co-supervisors
ESR1	Gemmi (IIT)	Dalcanale(UNIPR), Nicolopoulos(NMG), Brázda (FZU), Batuk (UA), Yu(THF)
ESR2	Gemmi (IIT)	Dalcanale(UNIPR), Xu (SU), Van Genderen (UBA), Stowasser (RCH), McMahon (IUCr)
ESR3	Hadermann (UA)	Kolb (JGU), Perez (DENS), David (NU)
ESR4	Palatinus (FZU)	Boullay(CNRS), Plaisier (EST), Müller (BASF), Hadermann (UA)
ESR5	Palatinus (FZU)	Gemmi (IIT), van der Wal (TSC), Gorelik (ULM), McMahon (IUCr)
ESR6	Kaiser(ULM)	Boullay(CNRS), Abrahams (UBA), Prangsma (ASI)
ESR7	Gorelik(ULM)	Mugnaioli (IIT), Steinfeld (ELD), Zou (SU)
ESR8	Kolb (JGU)	Van Genderen (UBA), Müller (BASF), Hadermann (UA)
ESR9	Kolb (JGU)	Palatinus (FZU), Steinfeld (ELD), Boullay (CNRS), McMahon(IUCr)
ESR10	Xu(SU)	Käck (AZ), Brázda (FZU), Zhang (eBIC), Abrahams (UBA), Yu(THF)
ESR11	Zou(SU)	Waterman(STFC), Mugnaioli (IIT), Norberg (AZ), Kolb (JGU)
ESR12	Boullay (CNRS)	David (NU), Guilmeau (CNRS), Palatinus (FZU), Kolb (JGU), van der Wal (TSC)
ESR13	Boullay (CNRS)	Hadermann(UA), Kaiser(ULM), Plaisier (EST), Séguier (CDX)
ESR14	Abrahams (UBA)	Xu (SU), Prangsma (ASI), Kaiser (ULM), Zhang (eBIC)
ESR15	Abrahams (UBA)	Gemmi (IIT), Zou (SU), Waterman (STFC), Stowasser (RCH)

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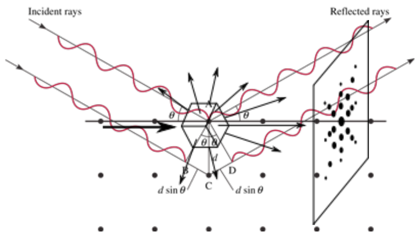
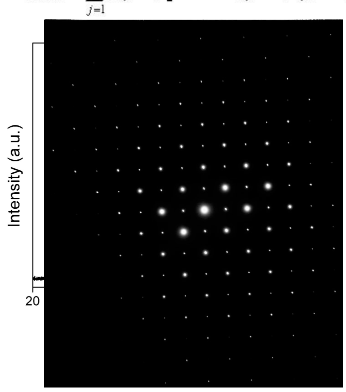
Diffraction pattern

Bragg's law: $n\lambda = 2d\sin\theta$

$$I\ \alpha\ |F|^2$$

$$F_{(h,k,l)} = \sum_{j=1}^{atoms} f_{(j)} \exp\left[2\pi \cdot i(hx_{(j)} + ky_{(j)} + lz_{(j)})\right]$$





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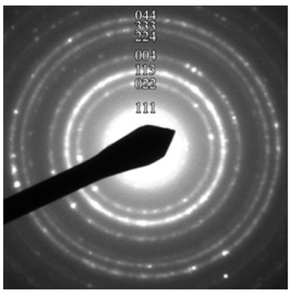
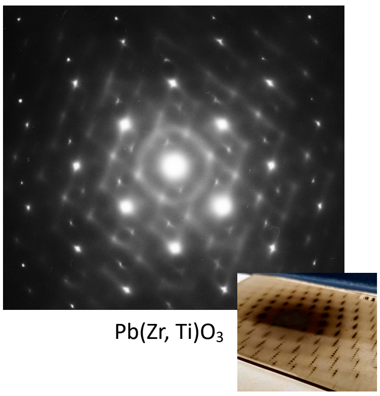
Electron diffraction pattern

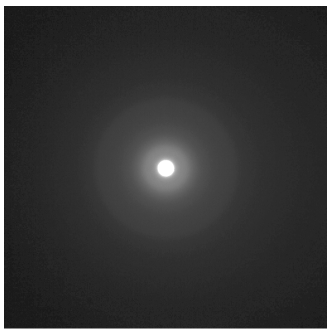
Long-range structure	Long- & medium-range structure	Short-range structure
Bragg spots	**Bragg spots & Diffuse scattering**	**Diffuse features & no Bragg spots**



Co oxide particles



$Pb(Zr, Ti)O_3$



Amorphous Si oxide

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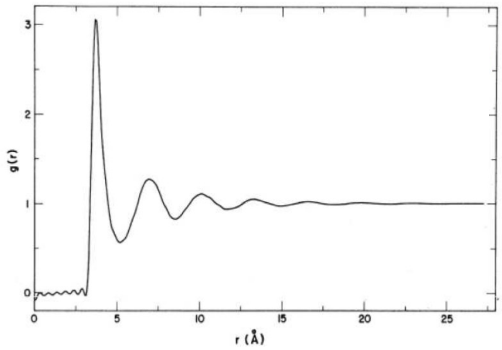
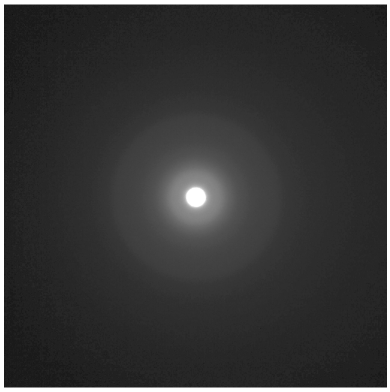
Diffraction and PDF

Real space information



Calculating PDF

Diffraction pattern
(Reciprocal space)



- Fourier transform of the scattering (scattered intensity)
- Histogram of inter-atomic distances

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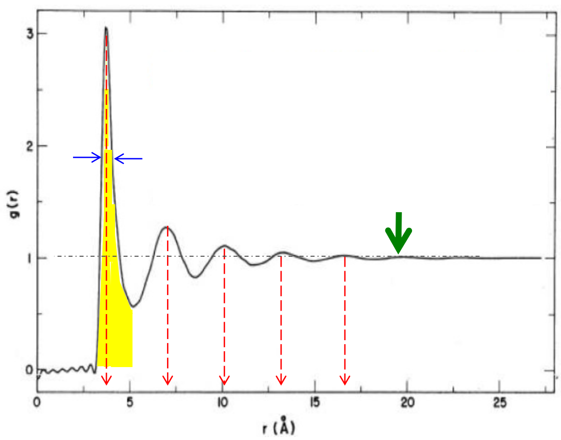
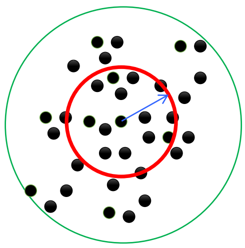
Pair distribution function

radial distribution function
(independent of orientation)





Peak position ⟶ atomic distance

Peak area ⟶ Coordination no.

Peak width ⟶ disorder (instrument)

R_{max} ⟶ particle size

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- Neutron
- Synchrotron x-ray

MAX IV and European Spallation Source (ESS) in Lund

- Electron

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Synchrotron & Neutron

- Large faculties
- Sample size:
  Meters -> ~100 nm
  Thick sample
- Q_{max} ~ 30-50Å^{-1}
- Relatively weak interaction
- Very good average overview of structure

Electron

- TEM in a lab
- Sample size:
  Few hundreds nm -> sub-nm
  Thin sample
- Q_{max} ~ 10-20Å^{-1}
- Strong interaction
- Versatility of microscope

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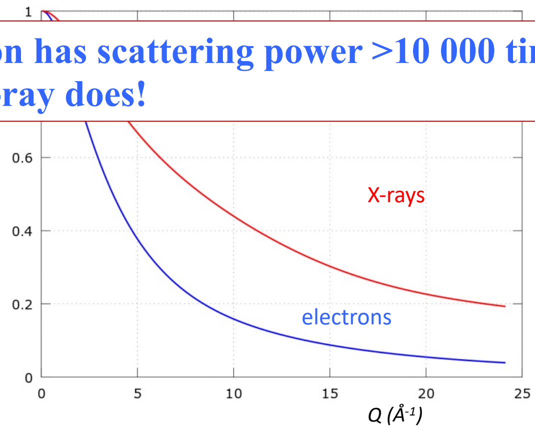
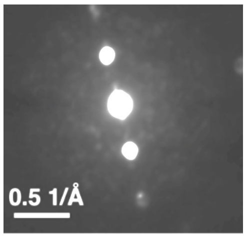
X-ray vs electron

Normalized scattering factors



Electron has scattering power >10 000 times stronger than x-ray does!



$$Q = 4\pi \sin\theta / \lambda$$

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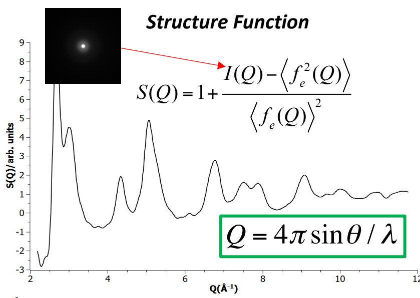
Work flow

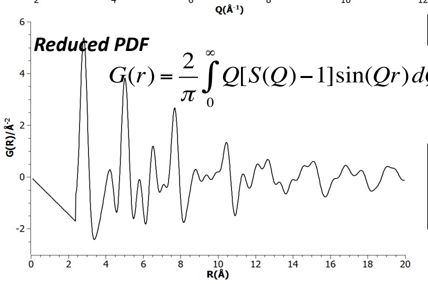
Structure Function



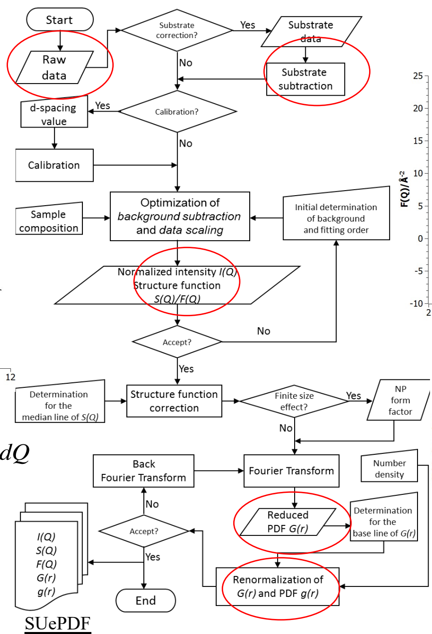
$$S(Q) = 1 + \frac{I(Q) - \langle f_e^2(Q) \rangle}{\langle f_e(Q) \rangle^2}$$

$$Q = 4\pi \sin\theta / \lambda$$

Reduced PDF

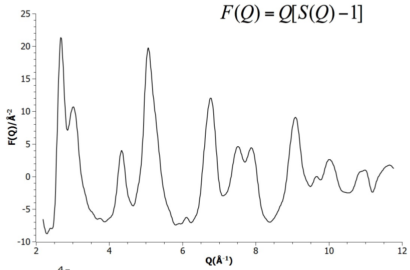


$$G(r) = \frac{2}{\pi} \int_0^\infty Q[S(Q) - 1] \sin(Qr)\, dQ$$



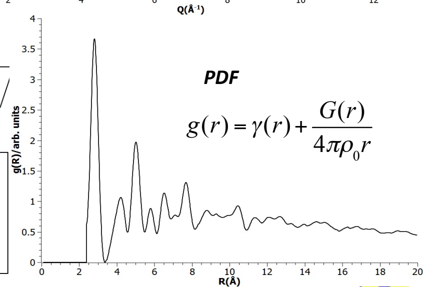
Reduced Structure Function

$$F(Q) = Q[S(Q) - 1]$$



PDF

$$g(r) = \gamma(r) + \frac{G(r)}{4\pi\rho_0 r}$$



SUePDF

Journal of Applied Crystallography, **50** (1), 304-312 (2017)

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Q-range

$$Q = 4\pi \sin\theta / \lambda$$

Higher Q; better results

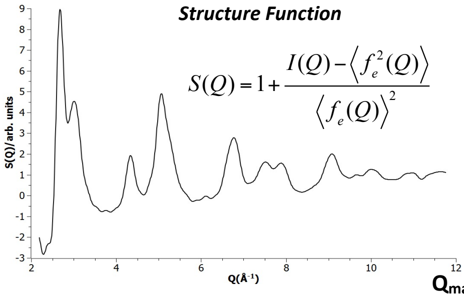
Laboratory-based:
Cu K_α X-ray: = 8 Å^{-1} (160°)
Mo = 17.4 Å^{-1}
Ag = 24 Å^{-1}

High-resolution X-ray: >20 Å^{-1}

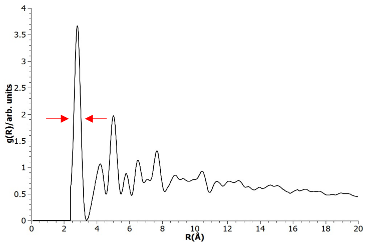
Neutron: >40 Å^{-1}

Structure Function

$$S(Q) = 1 + \frac{I(Q) - \langle f_e^2(Q) \rangle}{\langle f_e(Q) \rangle^2}$$



Q_{max}



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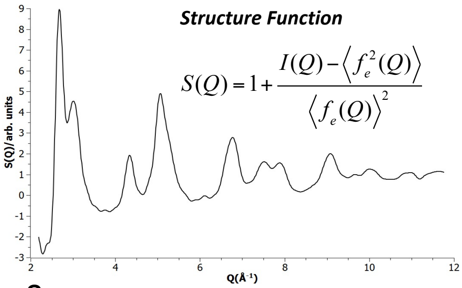
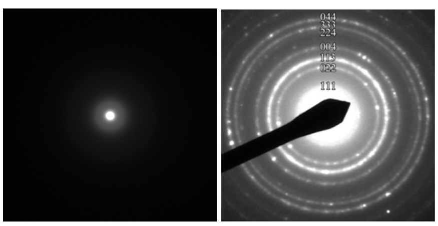
Q-range

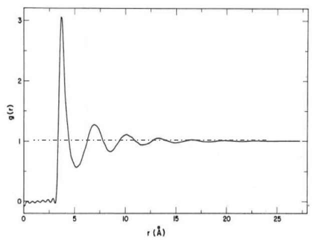
Structure Function

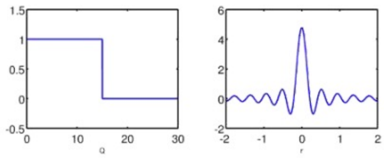
$$S(Q) = 1 + \frac{I(Q) - \langle f_e^2(Q) \rangle}{\langle f_e(Q) \rangle^2}$$



Q_{min} **Q_{max}**







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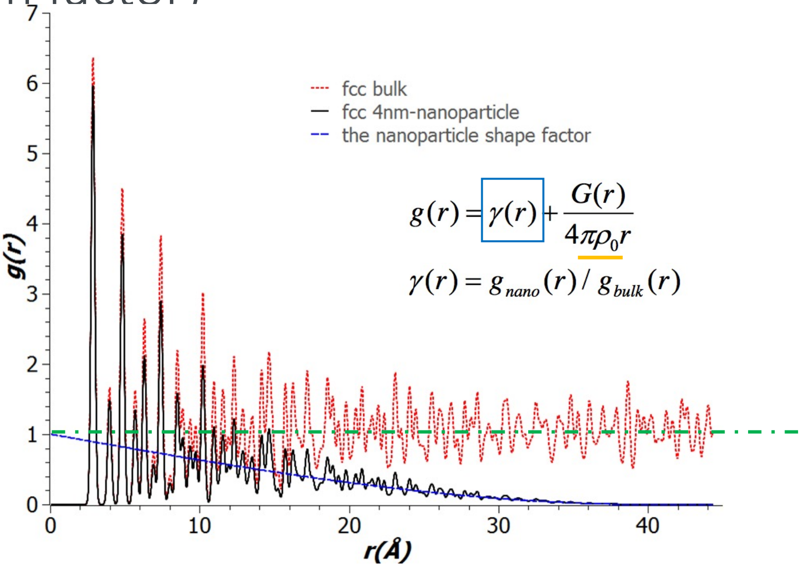
Shape (form factor)



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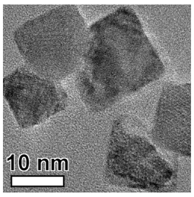
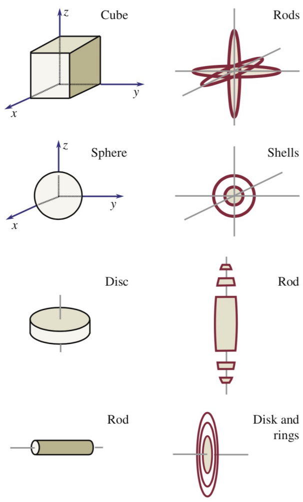
Shape (form factor)





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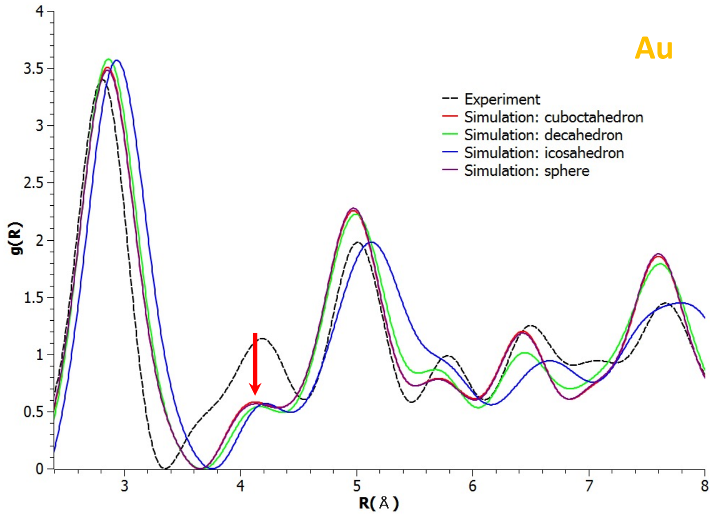
Shape (form factor)



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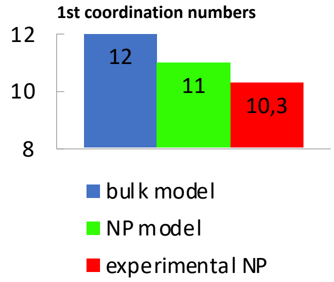
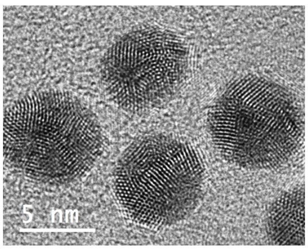
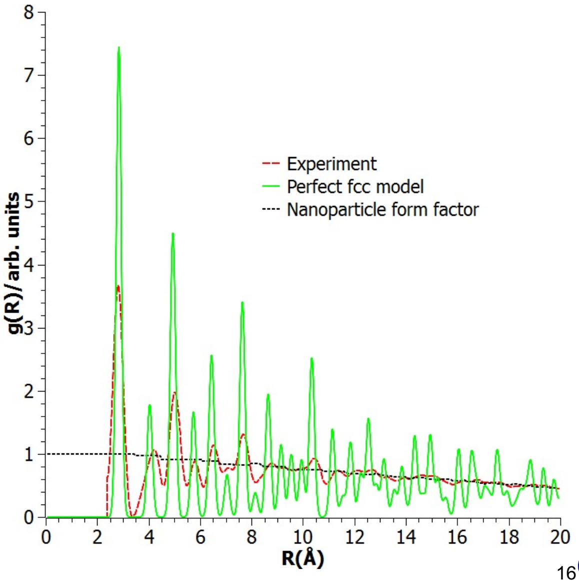
Au nanoparticles







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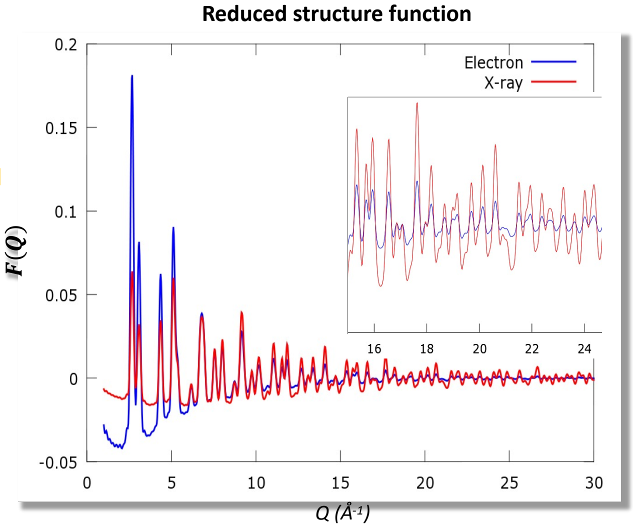
X-ray vs electron

Au



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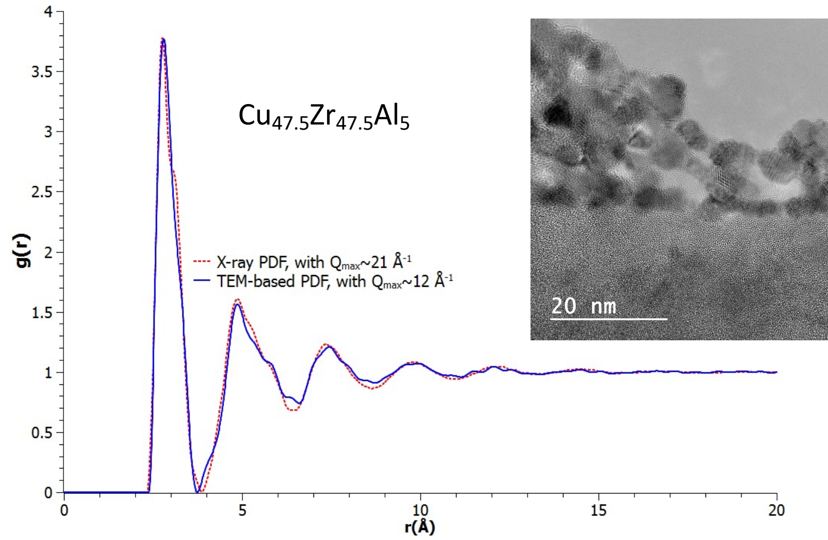
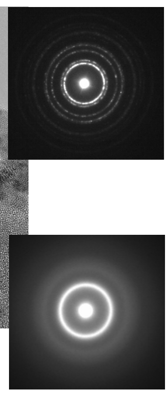
Metallic glass





*X-ray data from Ivan Kaban, Dresden (I. Kaban et al. Acta Mater 100 (2015) 369)

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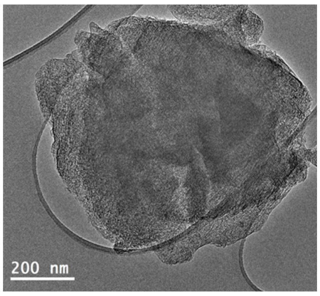
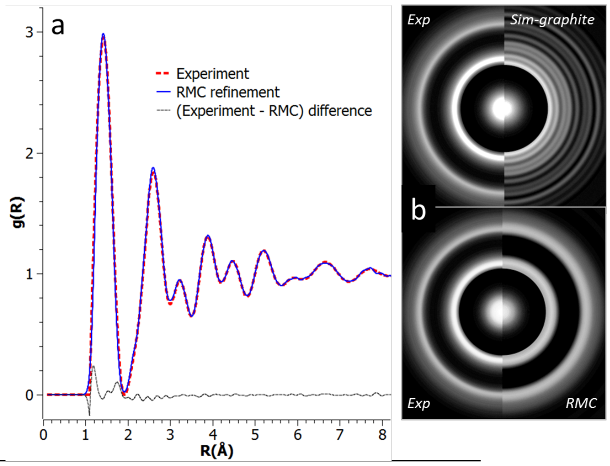
Nanoporous carbon





Amorphous-like nanoporous carbon

	TEM-based PDF	Graphite model (Fayos, 1999)	Neutron-based RMC (Zetterström et al., 2005)
1st peak position	1.42 Å	1.43 Å	1.40 Å
2nd peak position	2.60 Å	2.47 Å	2.45 Å
3rd peak position	3.22 Å	3.14 Å	2.82 Å
1st coordination number	2.51	3.00	2.54

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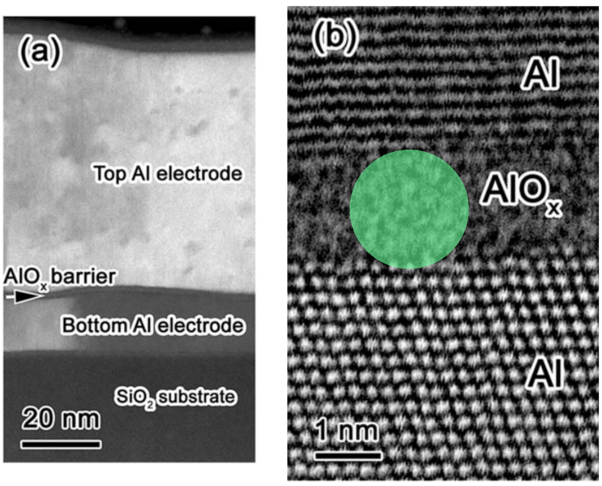
Al/AlO$_x$/Al Josephson junctions



Sci. Rep. 6 (2016) 29679

Collaboration with Dr. Lunjie Zeng & Prof. Eva Olsson at Chalmers University of Technology

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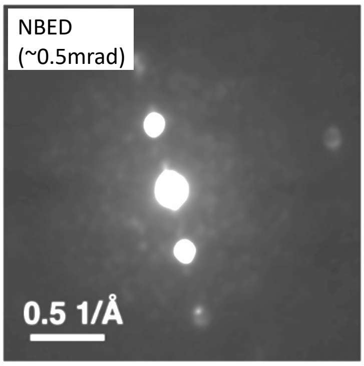

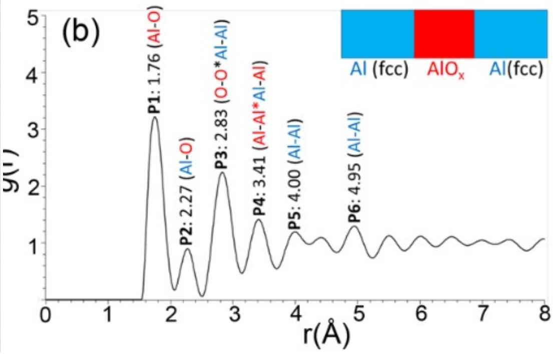

Q_{max} = 13 $Å^{-1}$

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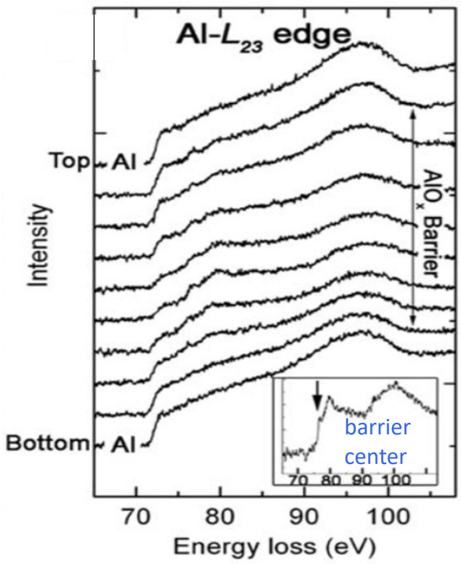

tetrahedral ($[AlO_4]^{-5}$)

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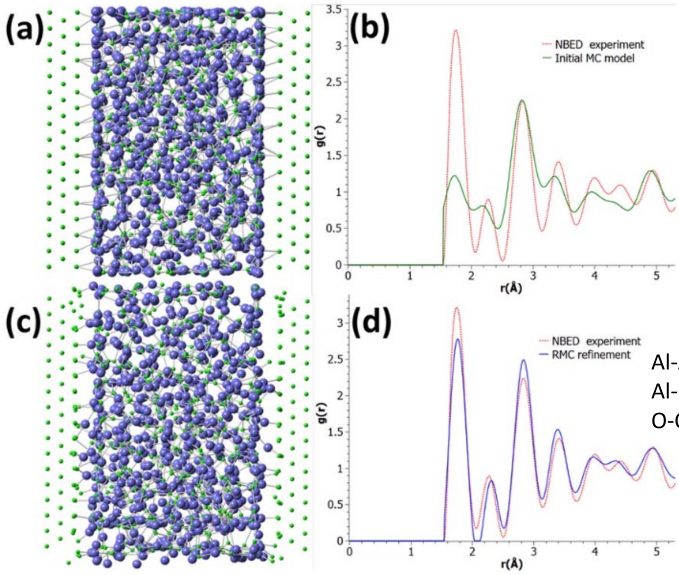
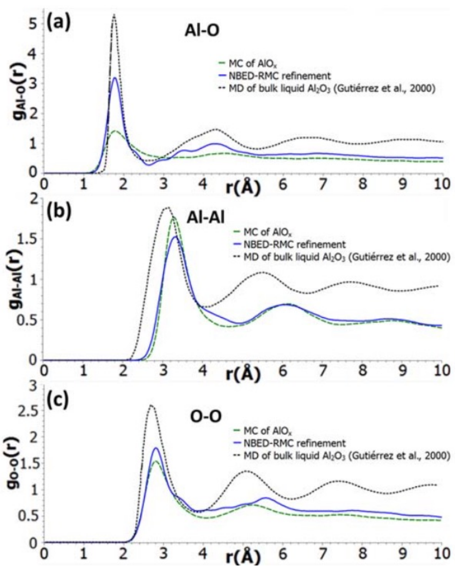
17× 27× 27 $Å^3$

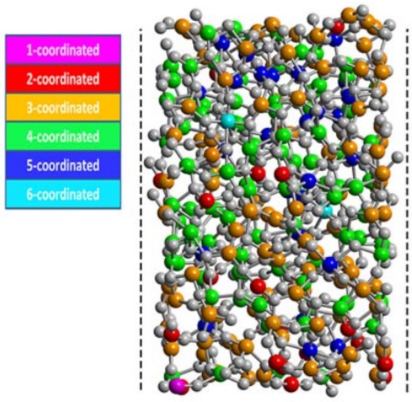
3.12 g/cm^3



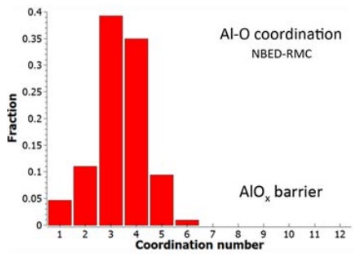
Al-Al: 3.1 Å
Al-O: 1.55 Å
O-O: 2.6 Å



Al atoms with different colors have different O (grey) coordination numbers.

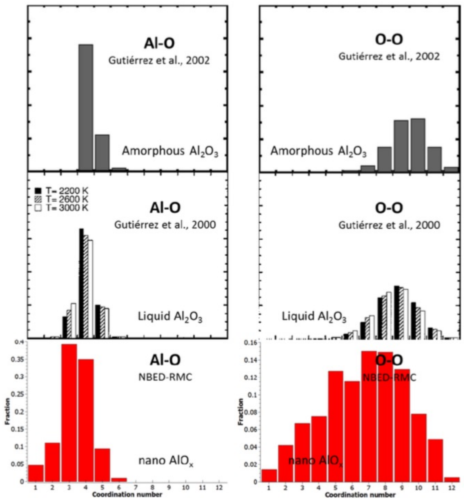


456 Al and 681 O atoms

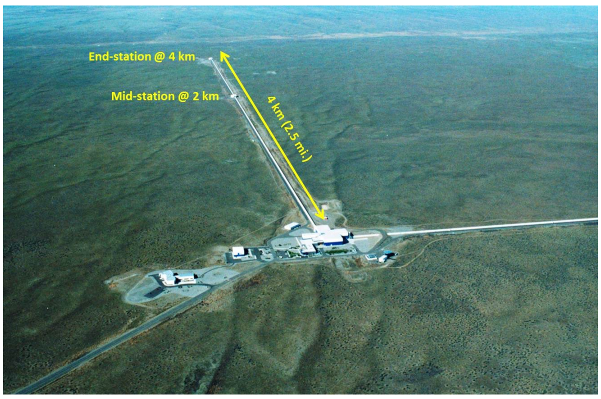


Model	Pair distance peak (Å)			Al-O coordination (%)			
	Al-O	Al-Al	O-O	3	4	5	6
NBED-RMC nano-AlO$_x$	1.77	3.31	2.81	39.2	34.9	9.4	0.9
MD Liquid bulk Al_2O_3*	1.75	3.15	2.75	13	66	20	≤1
MD Amorphous bulk Al_2O_3**	1.76	3.12	2.75	0	76	22	2
XRD amorphous bulk Al_2O_3***	1.80	3.20	2.80	20	56	22	0

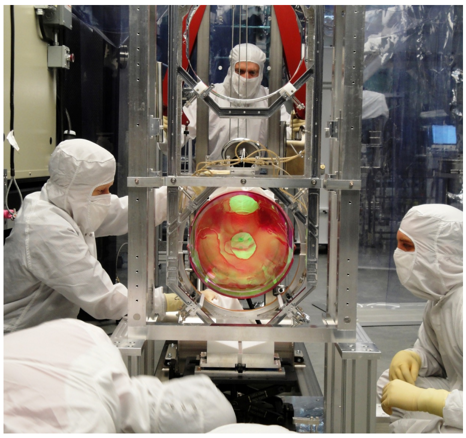
* Gutiérrez et al., Phys. Rev. E, 61, 2723 (2000).
** Gutiérrez et al., Phys. Rev. B, 65, 104202 (2002).
***P. Lamparter and R. Kniep, Physica B **234-236**, 405 (1997).



LIGO- Laser Interferometer Gravitational-Wave Observatory

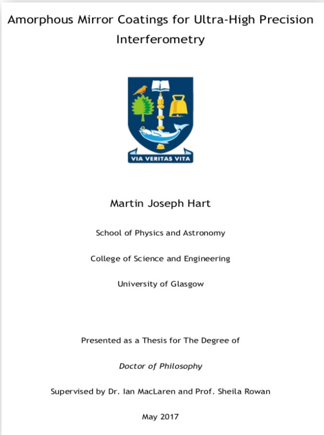


LIGO at Caltech,
https://www.ligo.caltech.edu/



https://www.ligo.org

LIGO



Amorphous Mirror Coatings for Ultra-High Precision Interferometry

VIA VERITAS VITA

Martin Joseph Hart

School of Physics and Astronomy

College of Science and Engineering

University of Glasgow

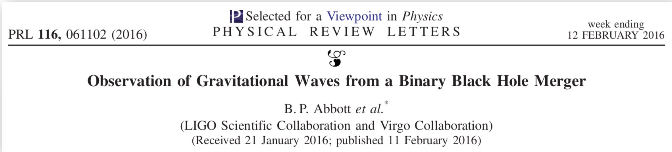
Presented as a Thesis for The Degree of

Doctor of Philosophy

Supervised by Dr. Ian MacLaren and Prof. Sheila Rowan

May 2017

Ti:Ta_2O_5 coating on glass



Selected for a Viewpoint in *Physics*

PRL **116,** 061102 (2016) PHYSICAL REVIEW LETTERS week ending 12 FEBRUARY 2016

Observation of Gravitational Waves from a Binary Black Hole Merger

B. P. Abbott *et al.*[*]

(LIGO Scientific Collaboration and Virgo Collaboration)

(Received 21 January 2016; published 11 February 2016)

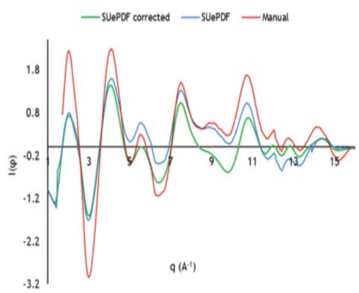


Figure 3-19 The improvements to the data obtained with SUePDF in comparison to performing the manual background subtraction.

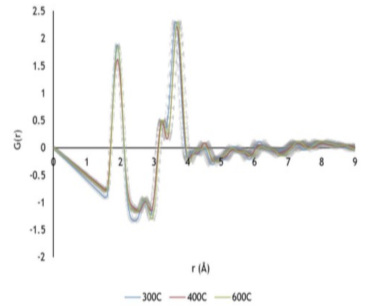


Figure 3-20 Reduced density functions (q = 16 Å⁻¹) of pure Ta_2O_5 annealed for 24 hours in air at 300˚C, 400˚C and 600˚C.

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Photocatalysts

COMMUNICATION

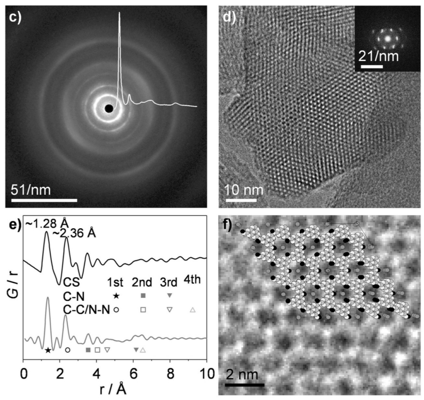
ADVANCED MATERIALS

Photocatalysis

www.advmat.de

"The Easier the Better" Preparation of Efficient Photocatalysts—Metastable Poly(heptazine imide) Salts

*Zupeng Chen, Aleksandr Savateev, Sergey Pronkin, Vasiliki Papaefthimiou, Christian Wolff, Marc Georg Willinger, Elena Willinger, Dieter Neher, Markus Antonietti, and Dariya Dontsova**



Adv. Mater. 29 (2017) 1700555

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Energy materials

Journal of Power Sources 511 (2021) 230444

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Crystallization behaviors in superionic conductor Na_3PS_4

Hiroshi Nakajima[a,*], Hirofumi Tsukasaki[a], Jiong Ding[a], Takuya Kimura[b], Takumi Nakano[b], Akira Nasu[b], Akihiko Hirata[c,d], Atsushi Sakuda[b], Akitoshi Hayashi[b], Shigeo Mori[a,**]

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[b] Department of Applied Chemistry, Osaka Prefecture University, Sakai, Osaka, 599-8531, Japan
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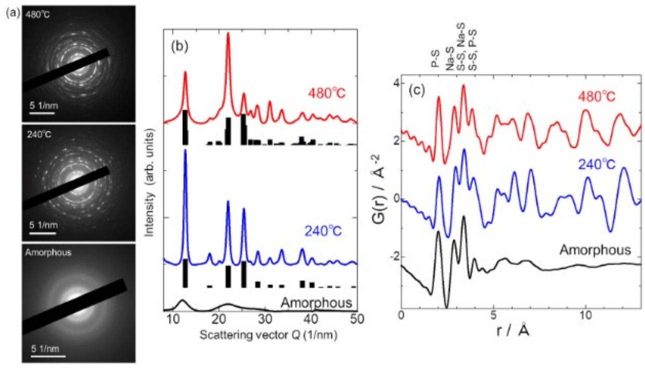


Amorphous (liquid-like) solids

nature materials

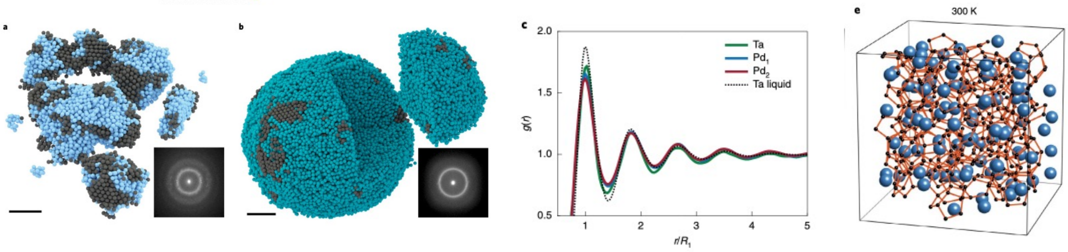
ARTICLES

https://doi.org/10.1038/s41563-021-01114-z

Check for updates

Three-dimensional atomic packing in amorphous solids with liquid-like structure

Yakun Yuan[1,5], Dennis S. Kim[1,5], Jihan Zhou[1,5], Dillan J. Chang[1], Fan Zhu[1], Yasutaka Nagaoka[2], Yao Yang[1], Minh Pham[3], Stanley J. Osher[3], Ou Chen[2], Peter Ercius[4], Andreas K. Schmid[4] and Jianwei Miao[1]✉



Nature Mater. (2021)
https://doi.org/10.1038/s41563-021-01114-z

Challenges

Synchrotron & Neutron

Data collection:
- Sample
- background
- ~~container~~

Electron

Data collection:
- Sample + background

Correction:
- Attenuation
- multiple scattering

Not easy for electron scattering (dynamical scattering)

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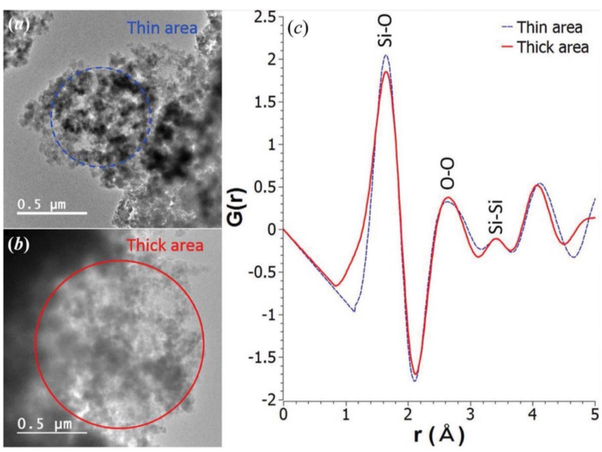
Thick regions

Amorphous silica

t/λ 0.7
2.5



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Intended study & Investigation

- ePDF slightly different to neutron and x-ray PDF
- “standard” routine
- background modeling and extract diffraction intensity
- ePDF calculation, including size and shape
- structural modeling or simulation (e.g. RMC)
- Applying to different materials
- Different conditions (heating, cooling, etc…)

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ePDF

STRUCTURAL SCIENCE
CRYSTAL ENGINEERING
MATERIALS
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Towards quantitative treatment of electron pair distribution function

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Available software packages for ePDF calculation from electron diffraction data.

Name	Includes diffraction data integration	Distribution
ProcessDiffraction†	Yes	Free
SUePDF (Tran *et al.*, 2017)	No	Free
eRDF Analyser (Shanmugam *et al.*, 2017)	Yes	Free
ePDF suite (NanoMegas, Belgium)	Yes	Commercial
ePDF tools (Shi *et al.*, 2019)	Yes	Free

† http://www.energia.mta.hu/~labar/ProcDif.htm

pyxem: https://github.com/pyxem/

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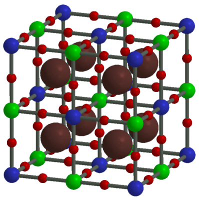
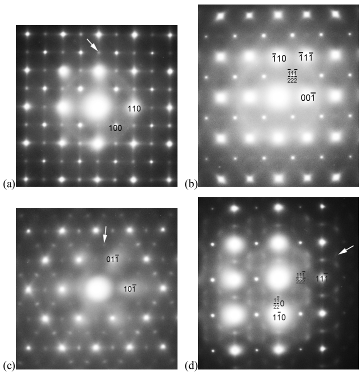
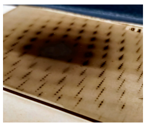
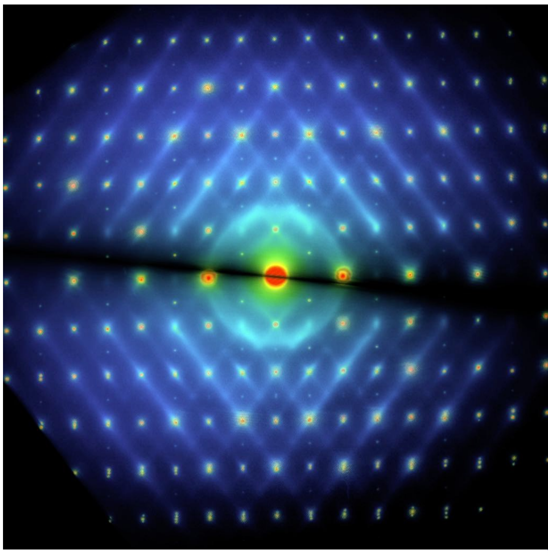
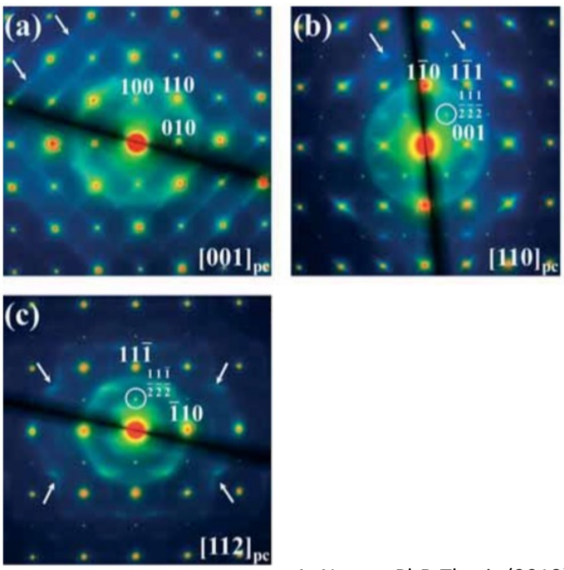
Pb-based perovskite ferroelectrics



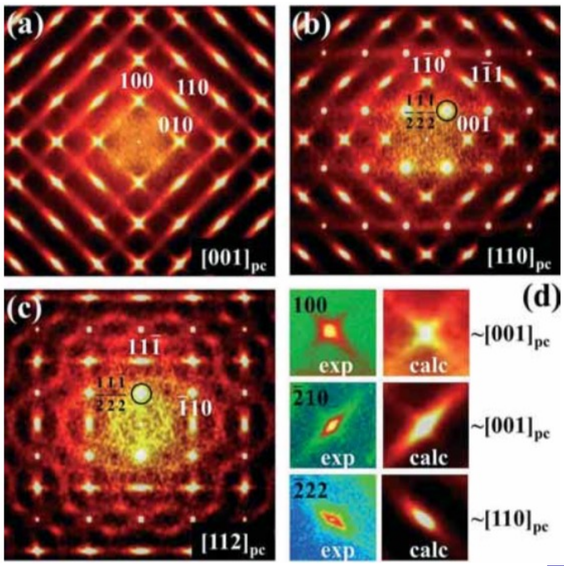








A. Neagu, PhD Thesis (2018)



Bragg reflections subtracted

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