OAK RIDGE National Laboratory

RMCProfile: Big Box modelling of crystalline to amorphous materials

Matt Tucker (ORNL, USA) Wojciech Slawinski (Warsaw)

ORNL is managed by UT-Battelle, LLC for the US Department of Energy

ADD 2026 Grenoble

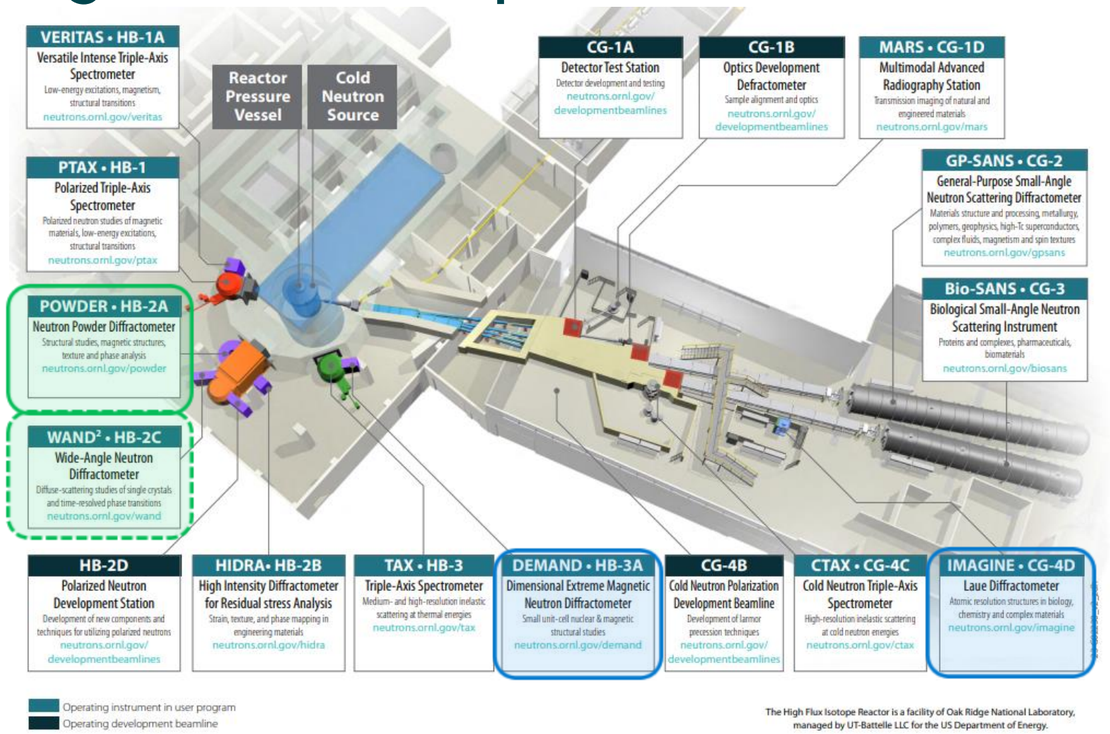
School and Conference on Analysis of Diffraction Data in Real Space

Diffraction Section @ ORNL

OAK RIDGE National Laboratory

Diffraction section includes 2 groups with 9 instruments

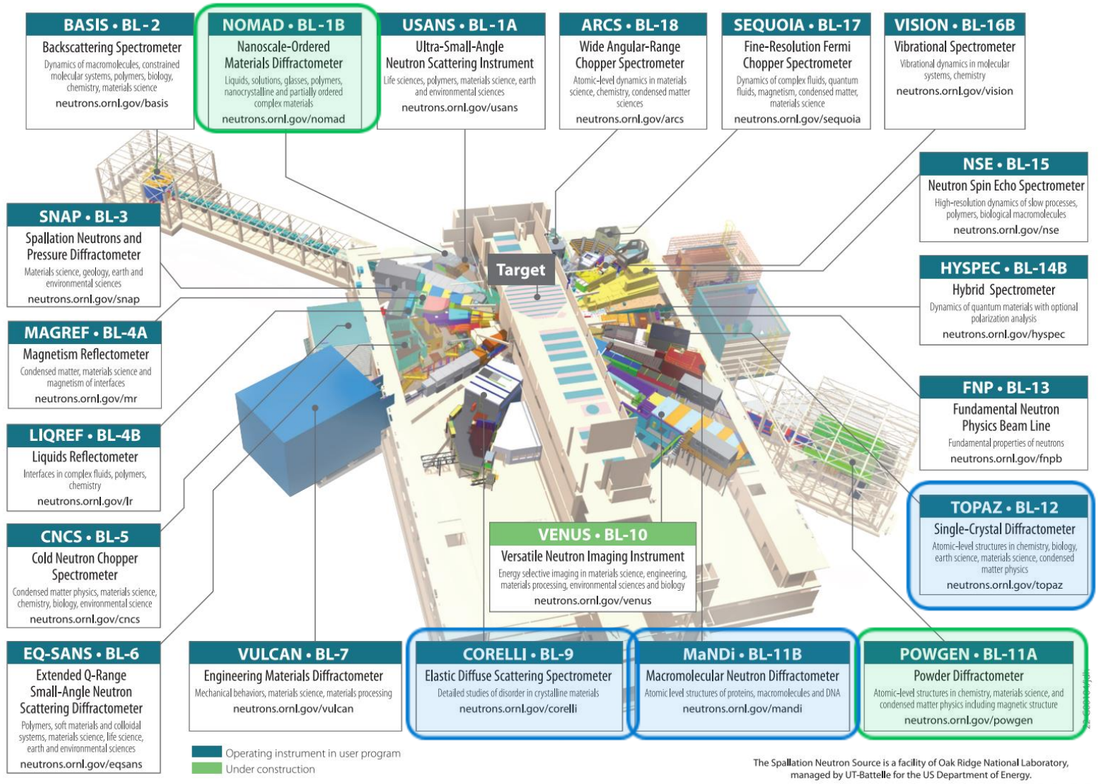
High Flux Isotope Reactor



- 2 Groups
  - Powder Diffraction Group
  - Single Crystal Diffraction Group

- 9 Instruments
  - 4 at the HFIR
  - 5 at the SNS

Spallation Neutron Source



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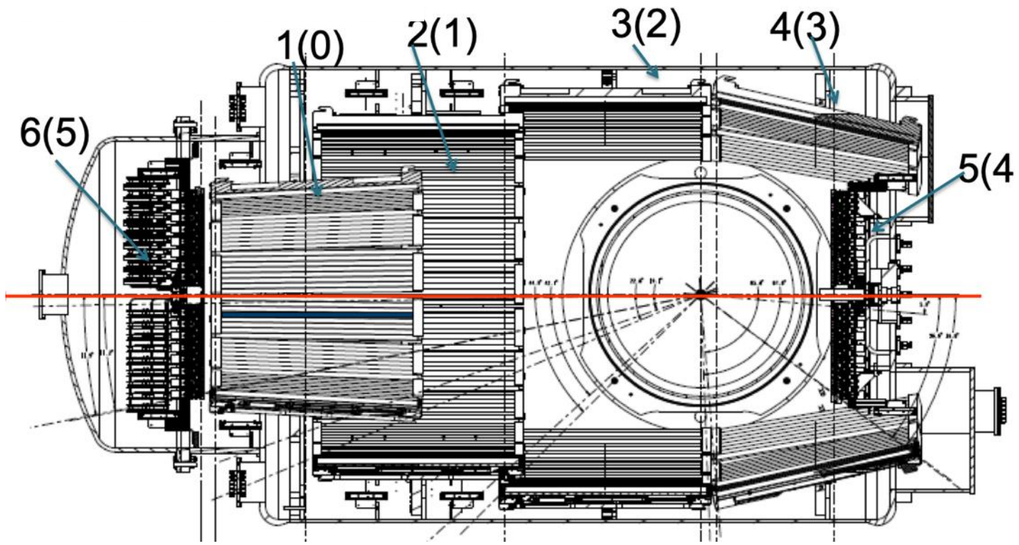
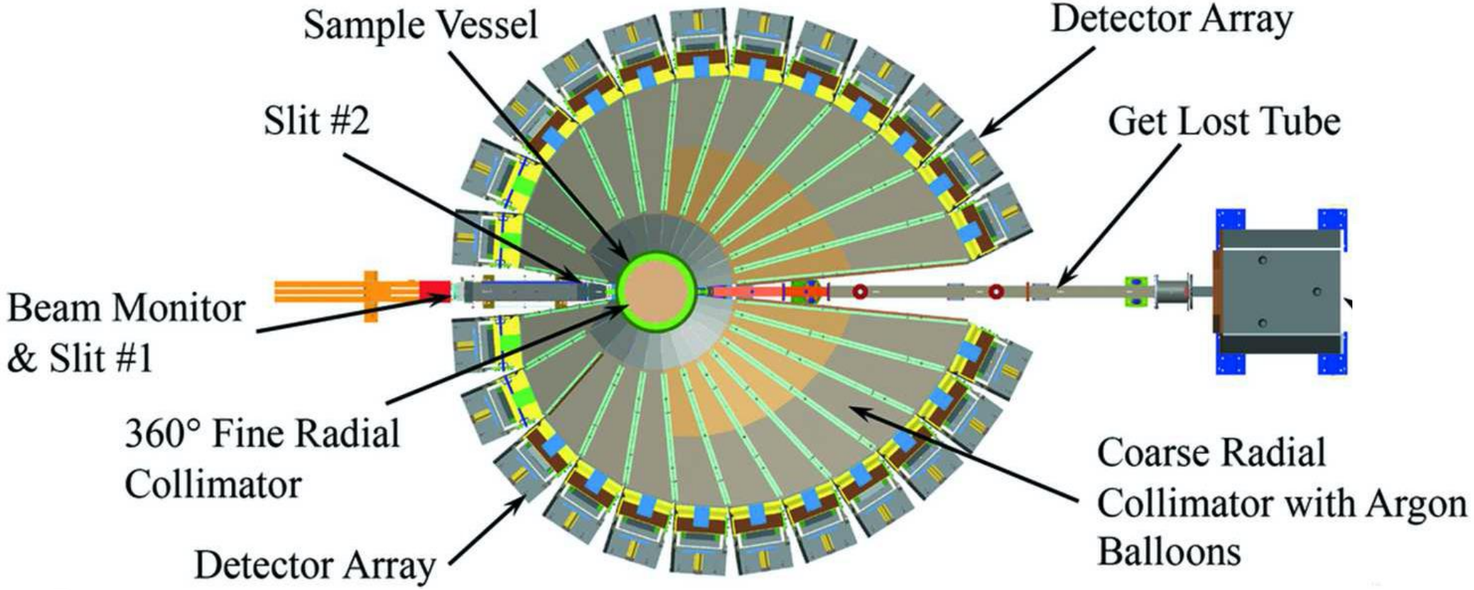
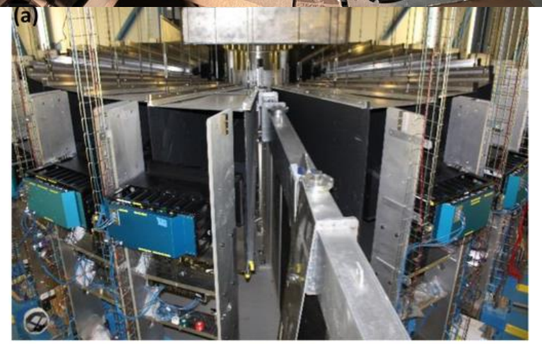
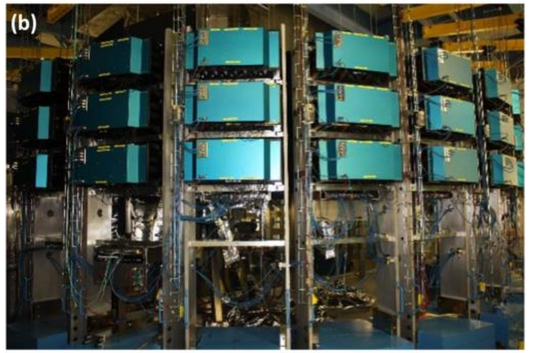
NOMAD and POWGEN









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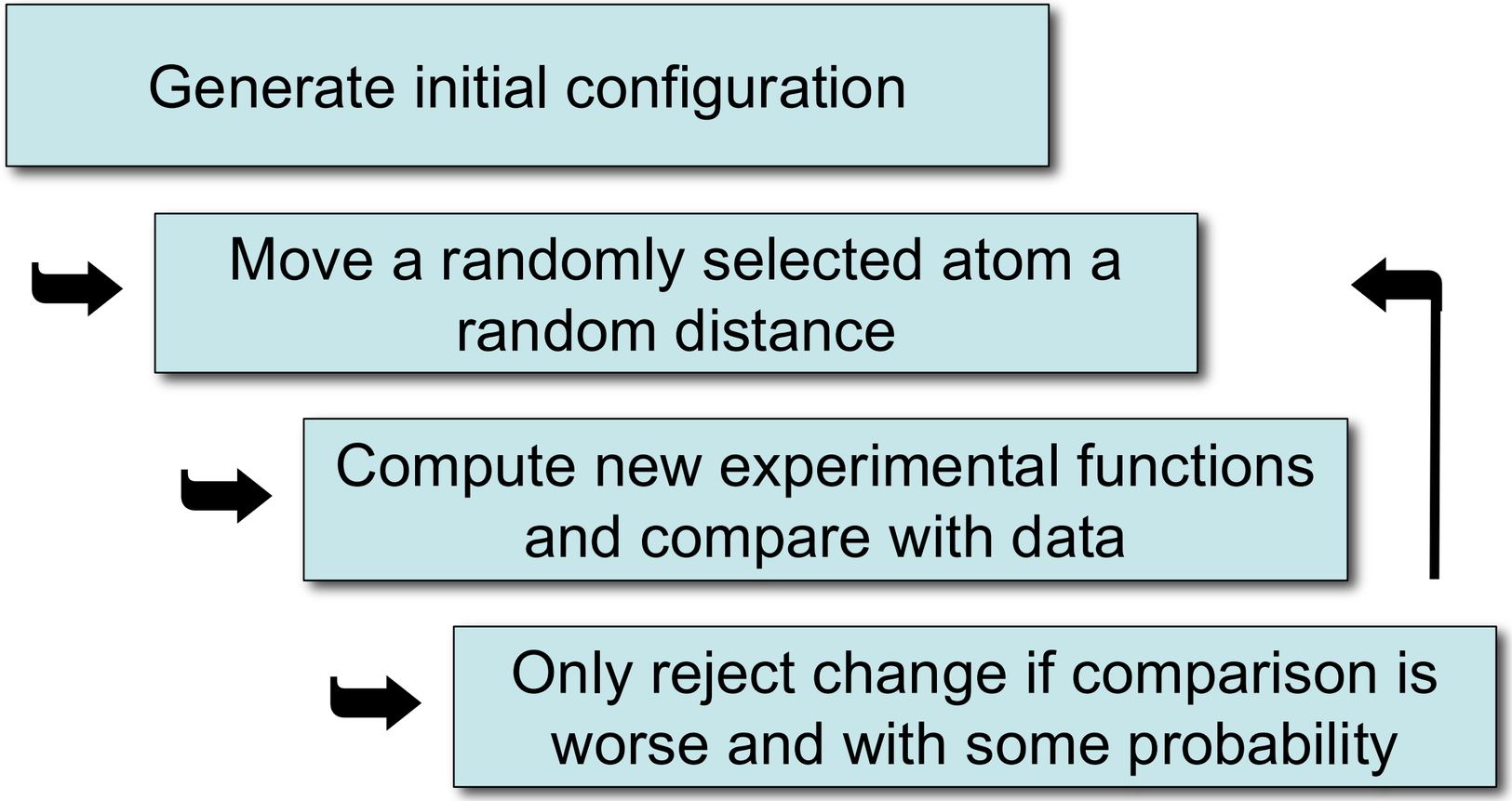
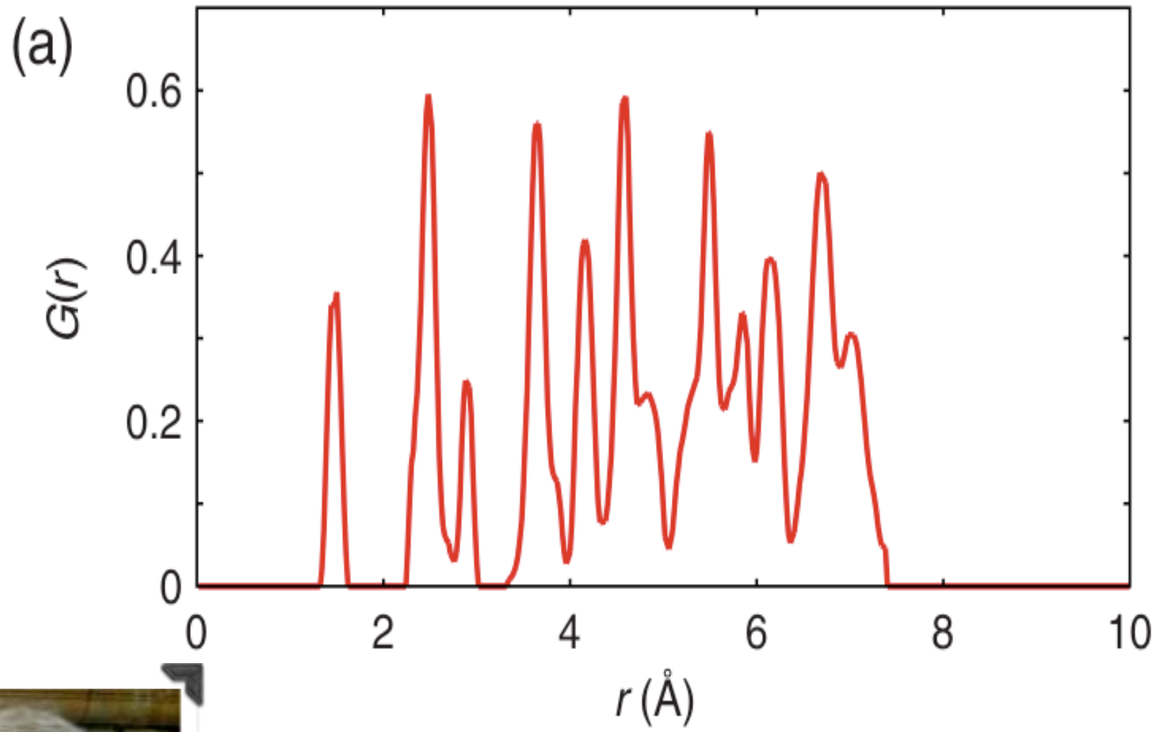
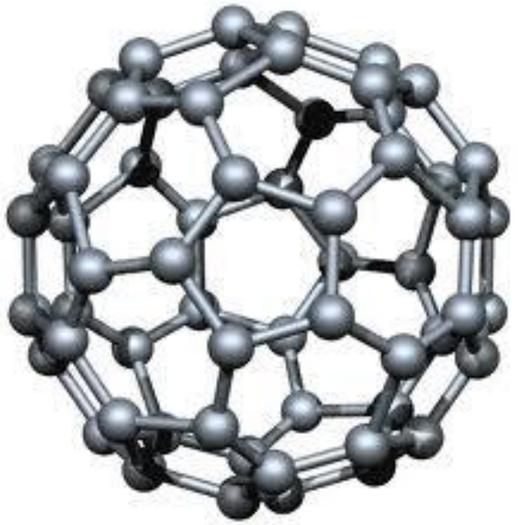
The Reverse Monte Carlo algorithm



RMC in action: C60





Cliffe M J *et al*
PRL **104** (2010) 1255013

INVERT

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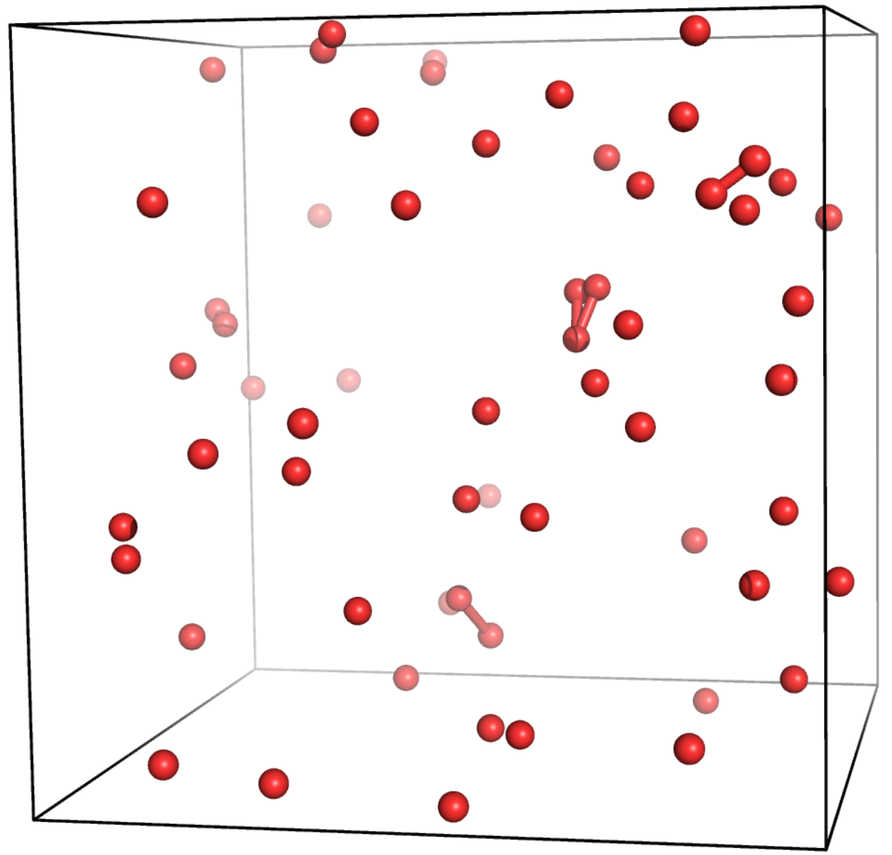
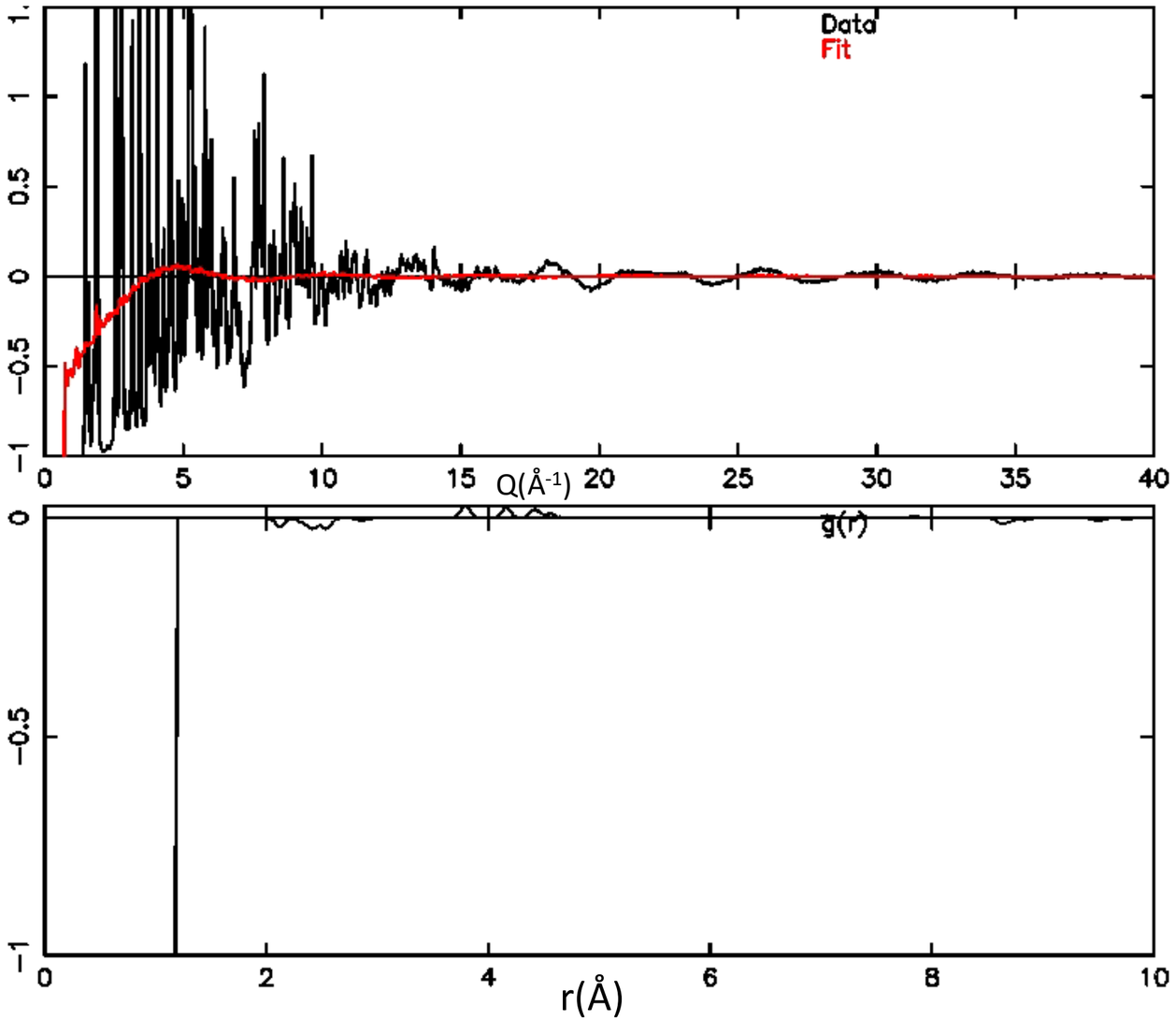
RMC in action: C60

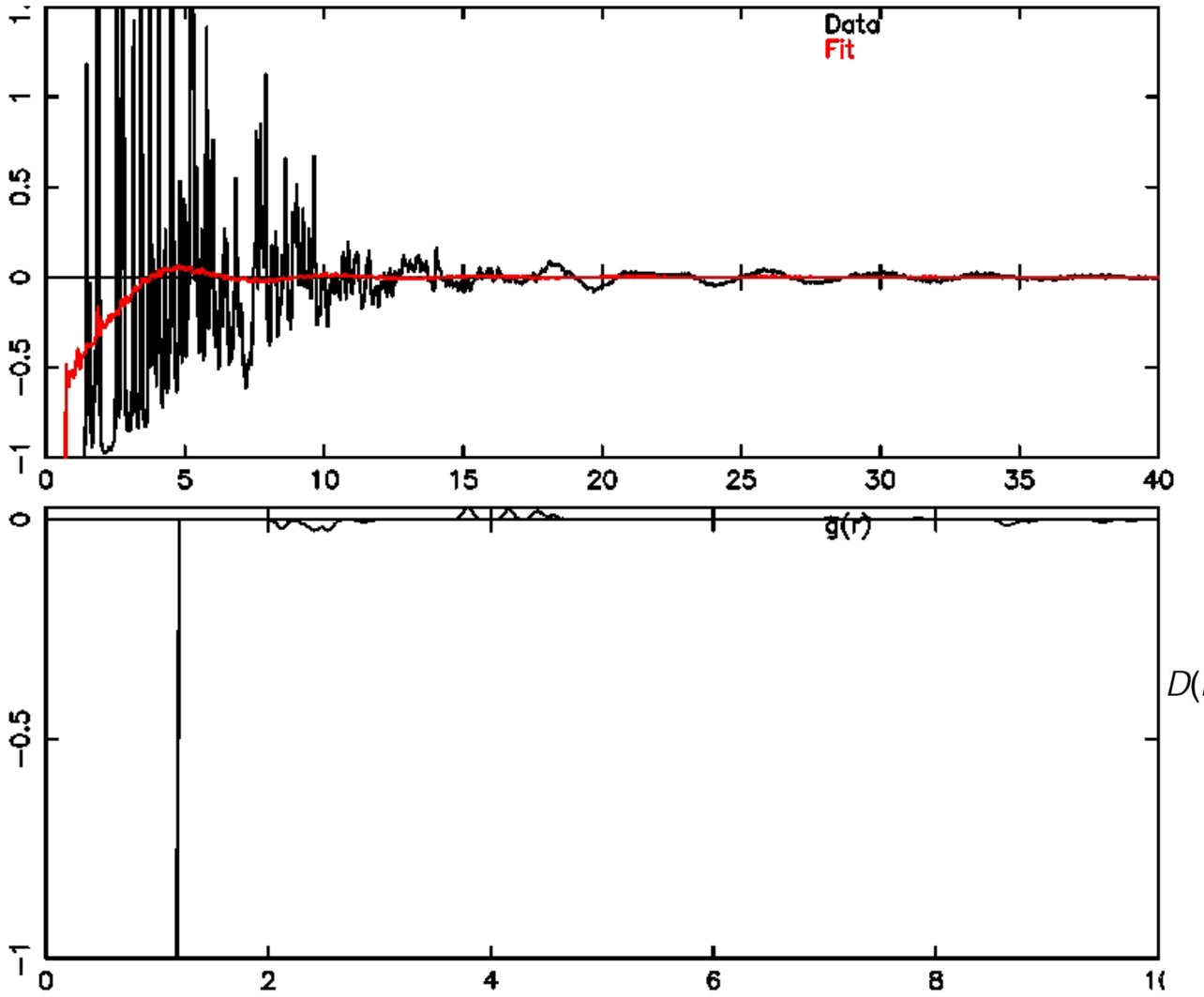


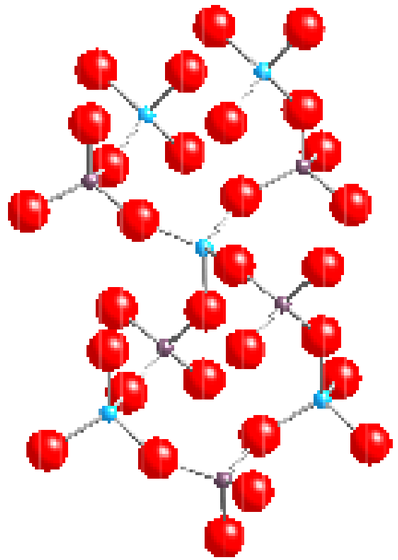
Cliffe M J *et al, PRL* **104** (2010) 1255013

RMC in action

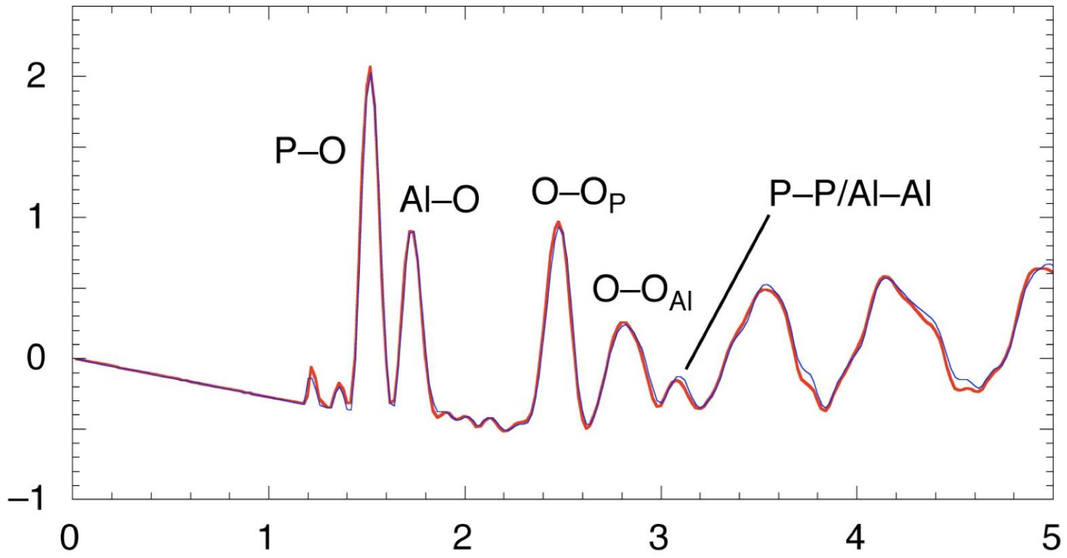


RMC in action





$AlPO_4$



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RMC programs

rmcprofile.pages.ornl.gov

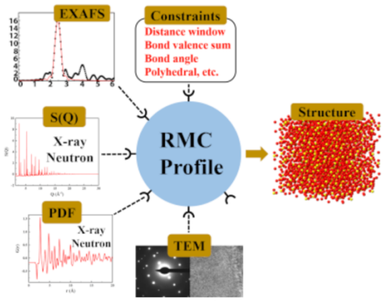
RMC PROFILE

SEARCH ABOUT POSTS PACKAGE COMMUNITY OTHERS

RMCProfile

Reverse Monte Carlo for crystalline and disordered materials

Welcome to the RMCProfile website hosted at Oak Ridge National Laboratory (ORNL) in US. Here you can download the RMCProfile software, view documentation and examples, join community for discussion and learn about updates of the package, etc.



This version of RMC was built from the original RMCA code of McGreevy & Pusztai to determine the local structure of crystalline materials while still being capable of analyzing disordered systems. The current version of RMCProfile results from a collaboration between scientists at ISIS facility (UK), Spallation Neutron Source (SNS at Oak Ridge National Laboratory, US), University of Cambridge (UK), University of Oxford (UK), Queen Mary University of London

https://rmcprofile.pages.ornl.gov/#

rmcprofile.ornl.gov

External Links

Monte Carlo

DISCUS
RMC++
HRMC
fullrmc
RMC for EXAFS
EPSR
RMC at ISIS facility
RMCProfile website - legacy
Wikipedia for RMC

RMC PROFILE

UT POSTS PACKAGE COMMUNITY OTHERS

ssion. Discussions are hosted as
ssion, one does need a GitHub

logy

UNIVERSITY OF OXFORD

Facilities

US - ORNL - NOMAD
US - ORNL - POWGEN
US - APS - 11-ID-B
US - BNL - 28-ID-1
UK - ISIS - GEM
UK - ISIS - Polaris
UK - Diamond - I15-1
Japan - J-PARC - NOVA
Japan - SPRING8 - BL14B1
Japan - SPRING8 - BL22XU
Europe - ESS - HEIMDAL
China - SSRF - BL13W1

Yuanpeng Zhang (ORNL)

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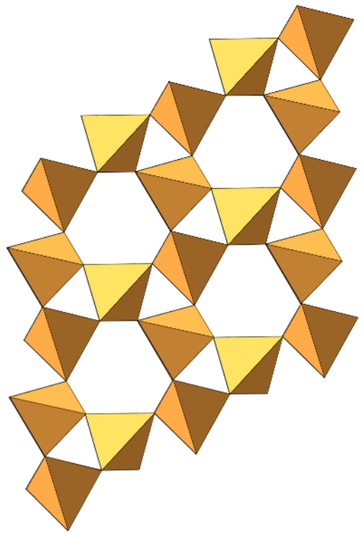
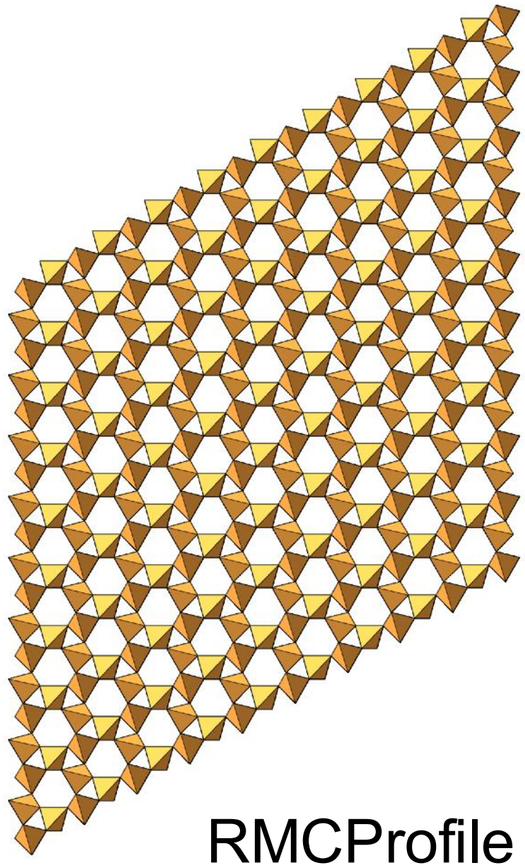
Big box vs small box models

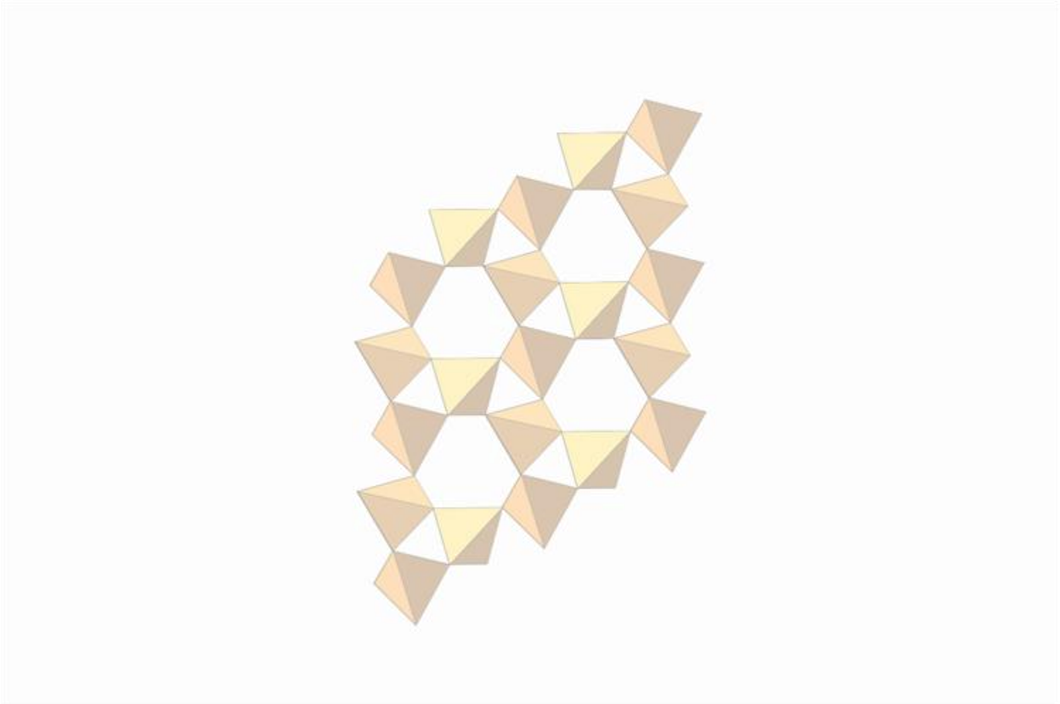


PDFgui
(r-space Reitveld)

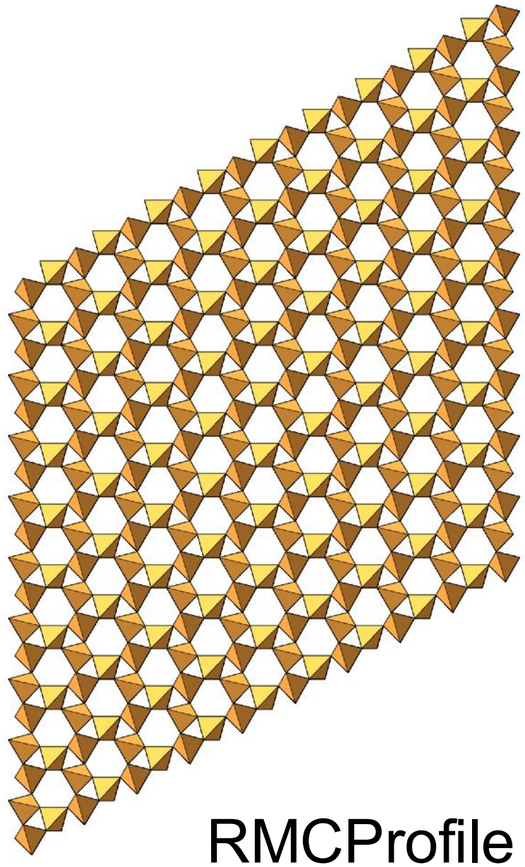


RMCProfile
(Reverse Monte Carlo)

Big box models



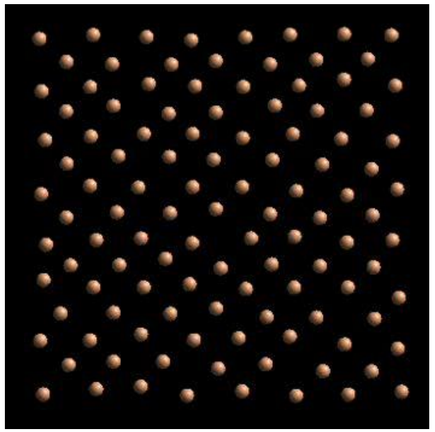
PDFgui
(r-space Reitveld)



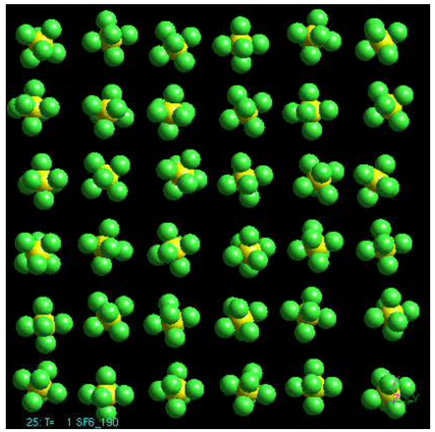
RMCProfile
(Reverse Monte Carlo)

Disordered materials

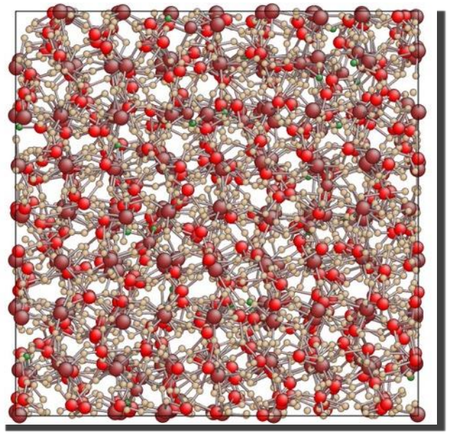
Simple crystals



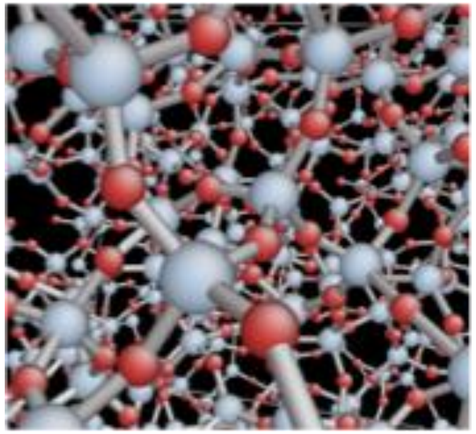
Disordered crystals



Amorphisation



Amorphous



RMCProfile

The RMC Method

Reverse Monte Carlo Simulation:
a new technique for the determination of disordered structures

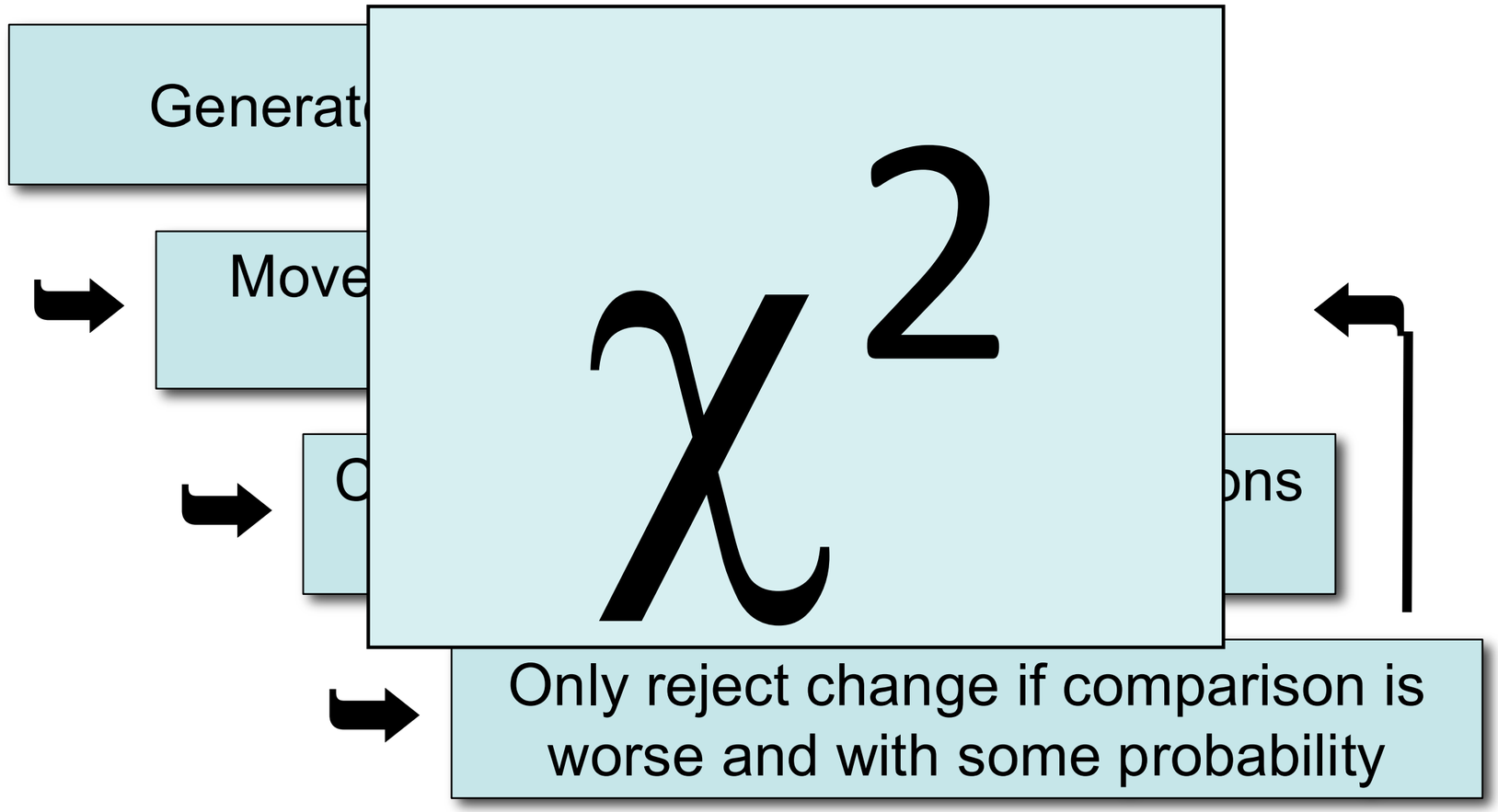
McGreevy R L and Pusztai L, *Molecular Simulation* **1**(1988) 359

We have developed a new technique, based on the standard Monte Carlo simulation method with Markov chain sampling, where a set of three dimensional particle configurations are generated that are consistent with the experimentally measured structure factor, A(Q), and radial distribution function, g(r), of a liquid or other disordered system. Consistency is determined by a standard χ^2 test using the experimental errors. No input potential is required. We present initial results for liquid argon. Since the technique can work directly from the structure factor it promises to be extremely powerful for modelling the structures of glasses or amorphous materials. It also has many other advantages in multicomponent systems and as a tool for experimental data analysis.

Key words: Monte Carlo, structure factor, radial distribution function, liquid, glass.

PACS numbers: 02.50, 61.25, 61.40.

The Reverse Monte Carlo algorithm



RMC χ^2

$$\chi^2_{RMC} = \sum_j \chi^2_j$$

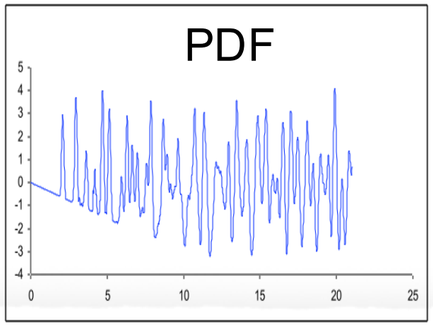
$$\chi^2_{F(Q)} = \sum_j \left[F_{calc}(Q_j) - F_{box}(Q_j)\right]^2 / \sigma^2_{F(Q)}(Q_j)$$ Total scattering

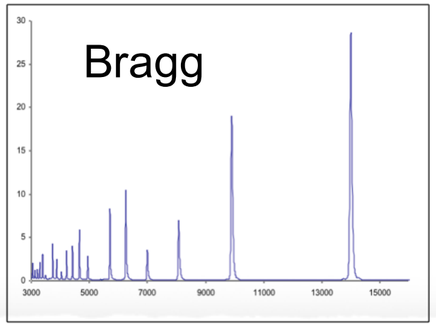
$$\chi^2_{G(r)} = \sum_j \left[G_{calc}(r_j) - G_{expt}(r_j)\right]^2 / \sigma^2_{G(r)}(r_j)$$ PDF

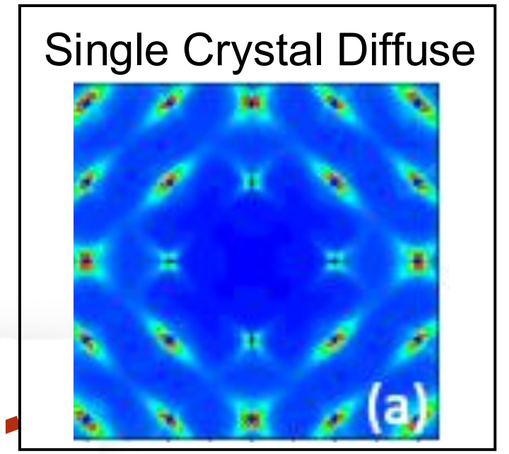
$$\chi^2_{profile} = \sum_j \left[I_{calc}(t_j) - s' I_{expt}(t_j)\right]^2 / \sigma^2_{I(i)}(t_j)$$ Bragg profile

$$\chi^2_f = \sum_k w_k \left[f_k^{calc} - f_k^{req}\right]^2$$ Constraints/restraints

RMC PROFILE







PRL 96, 047209 (2006) PHYSICAL REVIEW LETTERS week ending 3 FEBRUARY 2006

Magnetic Structure of MnO at 10 K from Total Neutron Scattering Data

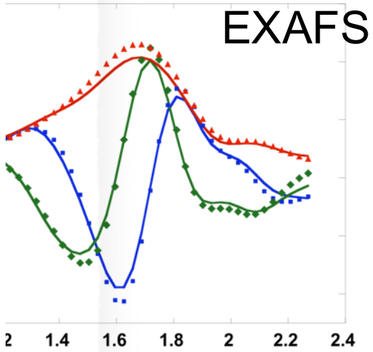
Andrew L. Goodwin,[1] Matthew G. Tucker,[1] Martin T. Dove,[1,*] and David A. Keen[2,3]

[1]*Department of Earth Sciences, Cambridge University, Downing Street, Cambridge CB2 3EQ, United Kingdom*
[2]*Department of Physics, Oxford University, Clarendon Laboratory, Parks Road, Oxford OX1 3PU, United Kingdom*
[3]*ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire OX11 0QX, United Kingdom*

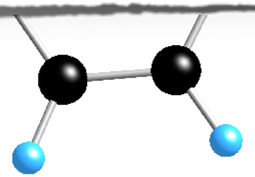
(Received 24 October 2005; published 2 February 2006)

Total neutron scattering data from a powdered sample of MnO collected at 10 K have been analyzed using the reverse Monte Carlo method to refine the nuclear and magnetic structure. The results give the first unambiguous assignment of the average magnetic structure. The magnetic moments are aligned ferromagnetically within (111) sheets with the magnetization vectors of alternate sheets along axes parallel and antiparallel to the $\langle 11\bar{2} \rangle$ directions, albeit with a small modulated out-of-plane component. Small displacements of Mn and O (modulated with the same periodicity) accompany the magnetic ordering and both atomic and magnetic structures may be described in the monoclinic space group $C2$.

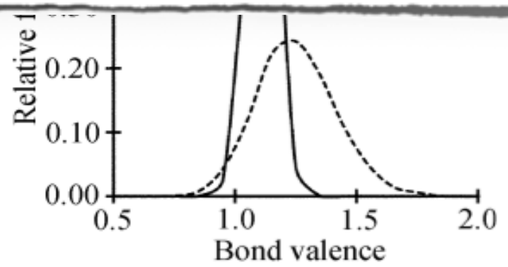
DOI: 10.1103/PhysRevLett.96.047209 PACS numbers: 75.25.+z, 02.70.Uu, 61.12.Ex

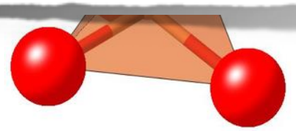


Polyhedral restraints



Molecular potentials





M G Tucker et al, *J. Phys.: Condens. Matter* **19,** 335218 (2007).

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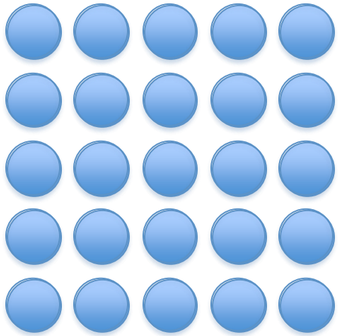
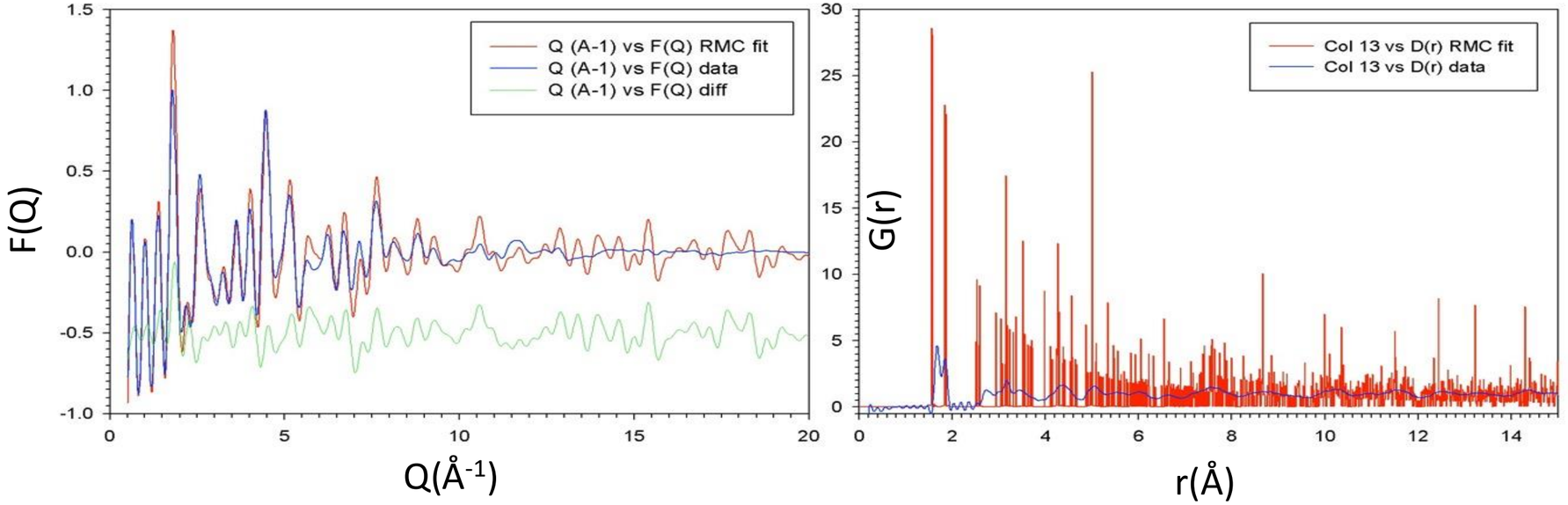
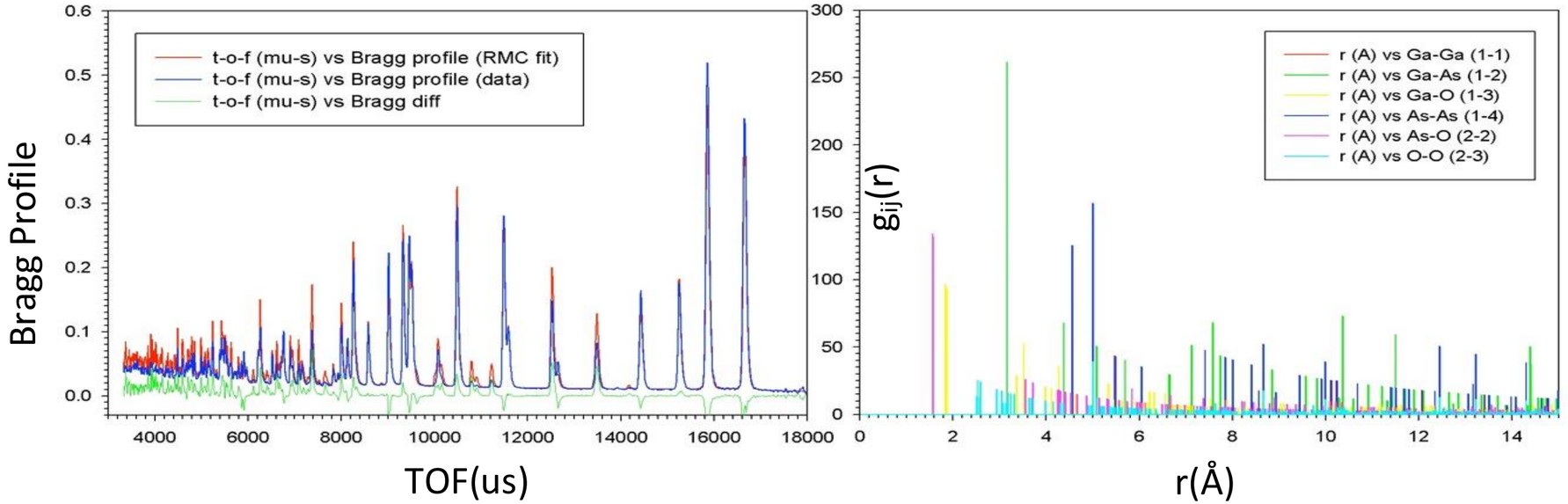
RMCProfile refinement



GaAsO4 RT.files





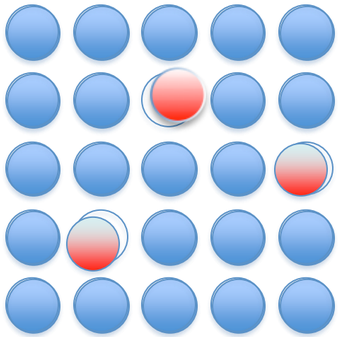
4608 atoms

0 moves

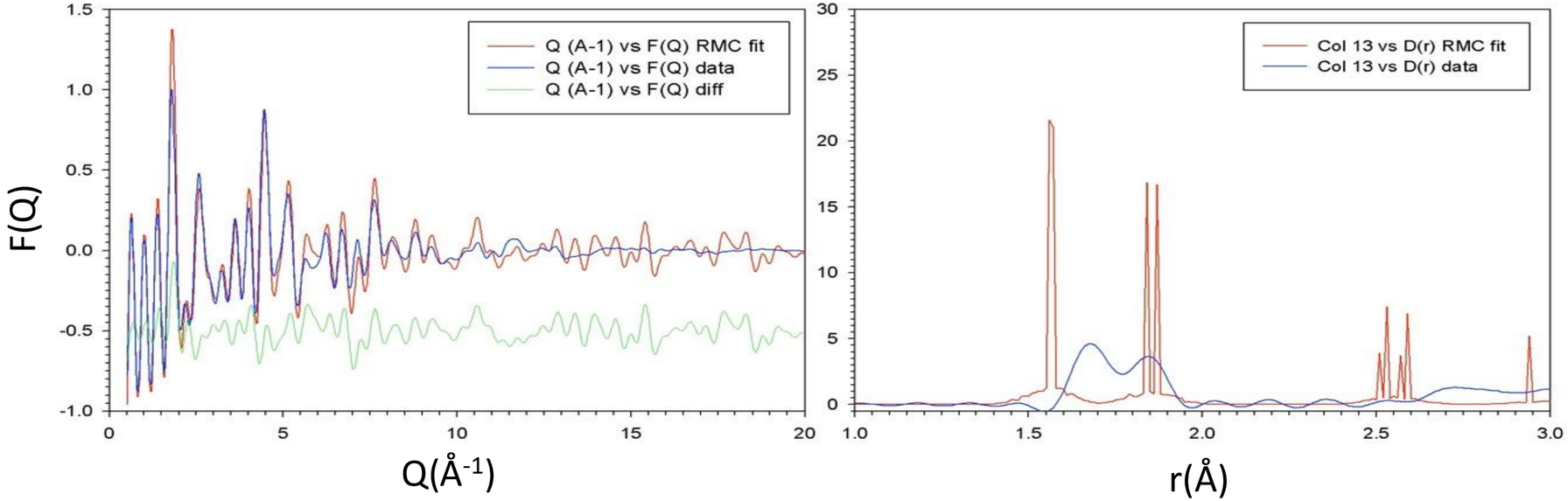
$\chi^2 = 3023$

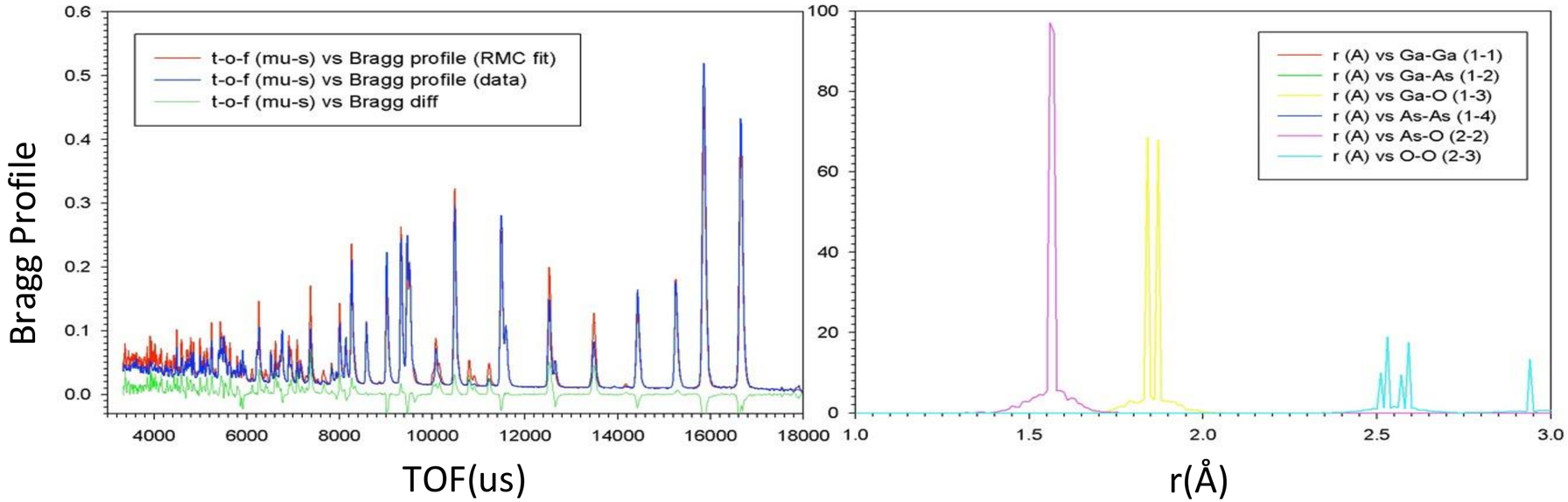
Slides from
Dave Keen

RMCProfile refinement



GaAsO4 RT.files





4608 atoms

834 moves

$\chi^2 = 1629$

Slides from
Dave Keen

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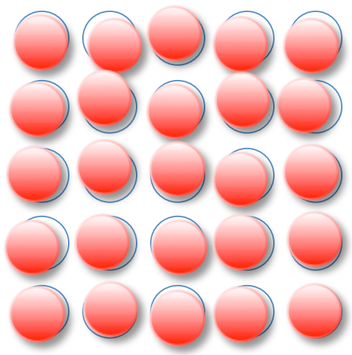
RMCProfile refinement

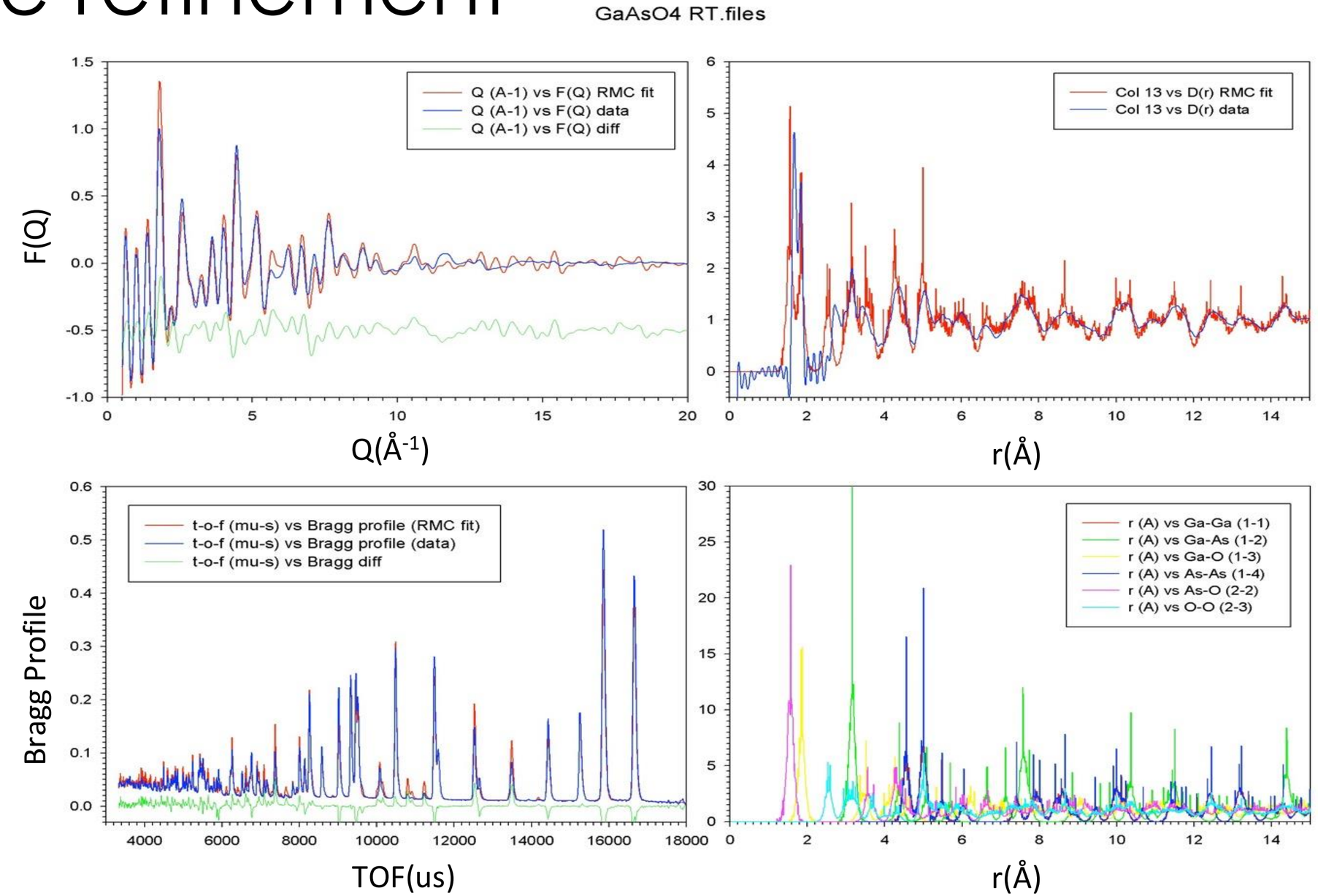


4608 atoms

4699 moves

$\chi^2 = 92.3$

Slides from
Dave Keen



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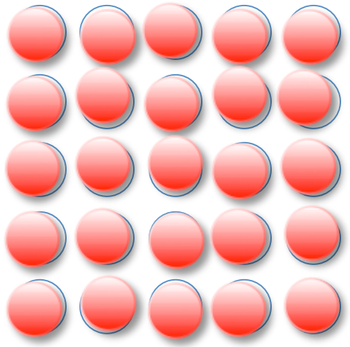
RMCProfile refinement

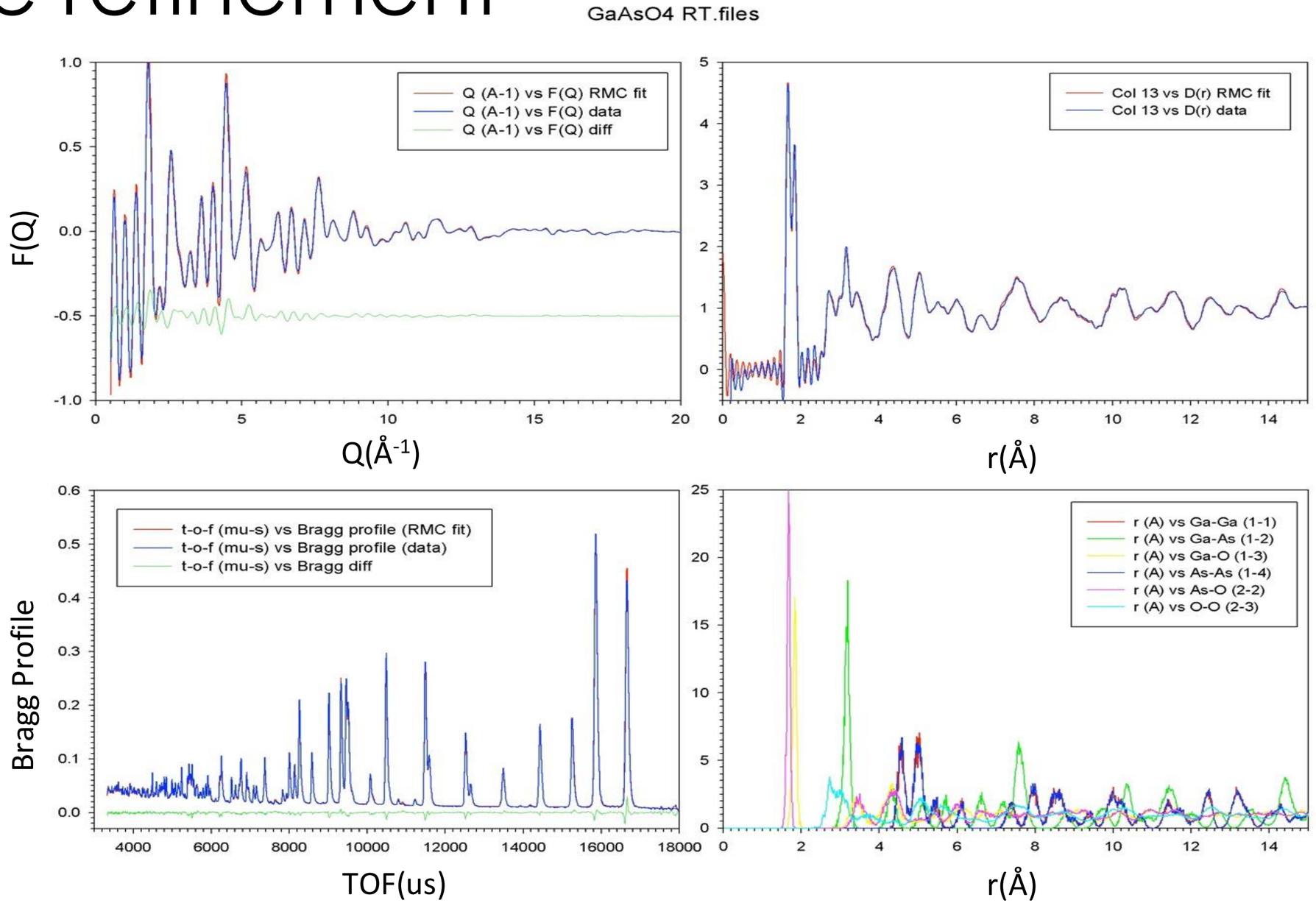


4608 atoms

RMC converged

$\chi^2 = 3.43$

Slides from
Dave Keen



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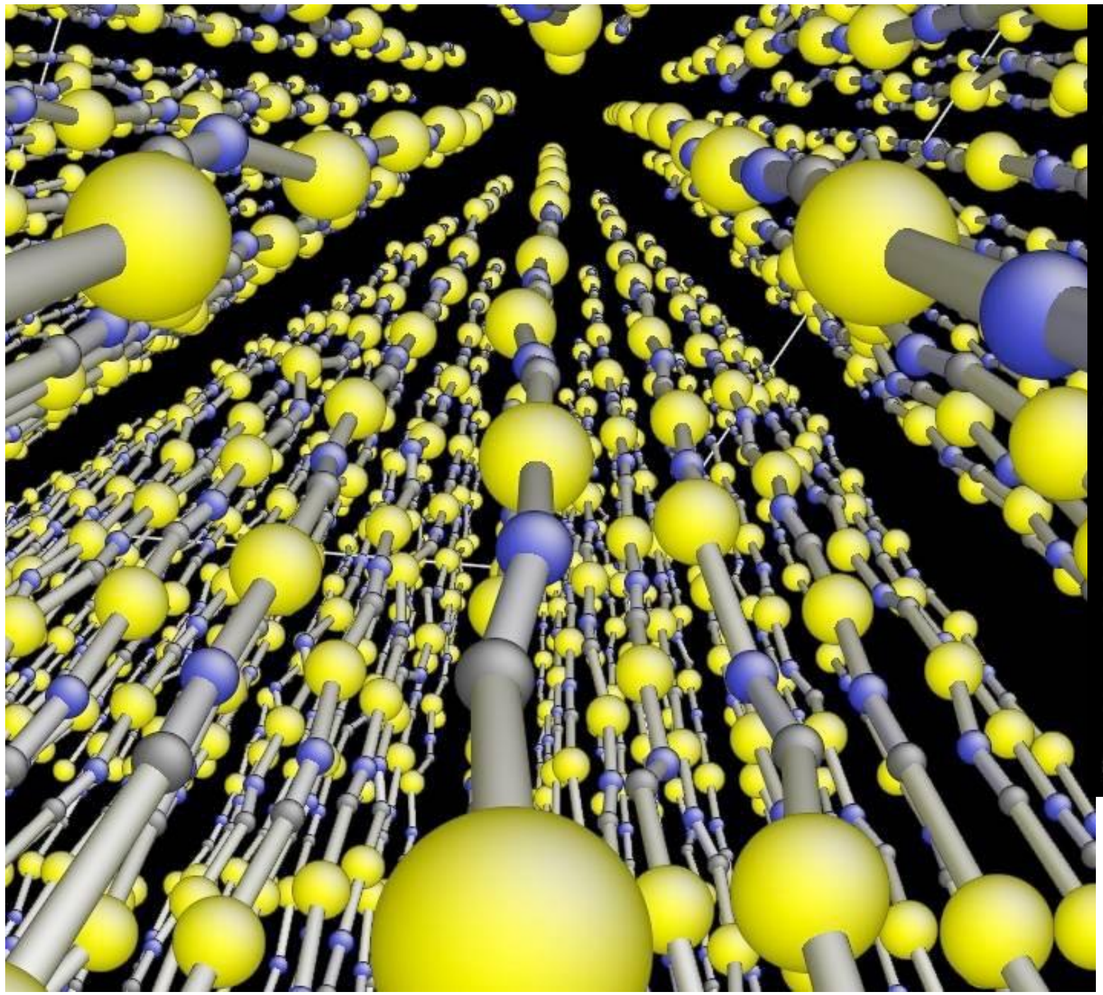
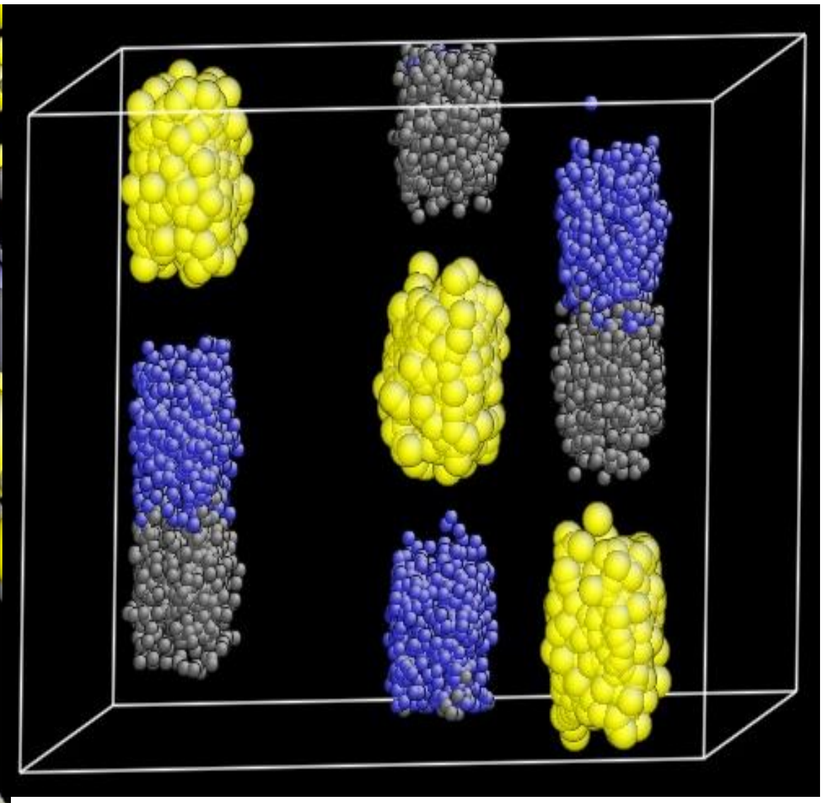
Big box models





Collapsed to unit cell

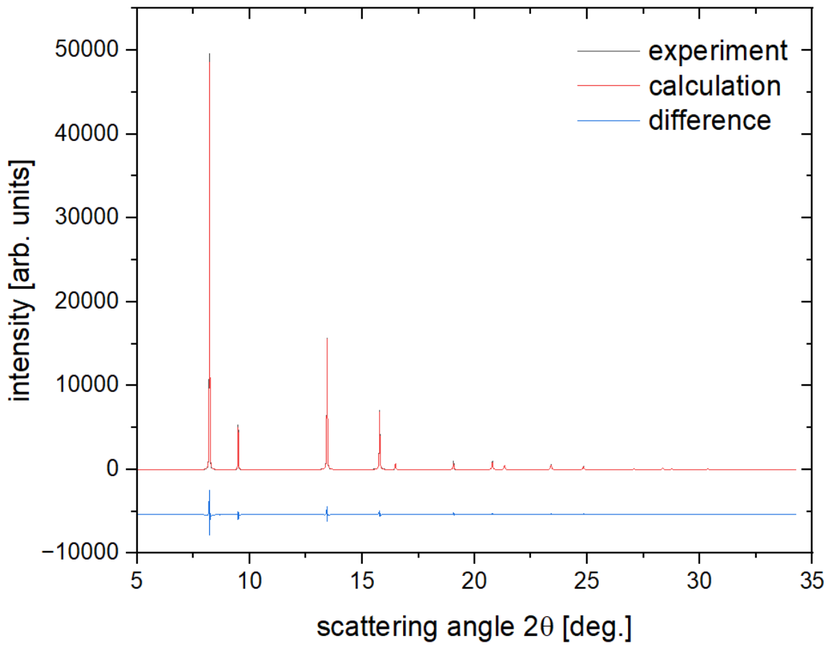
RMCProfile7

- **Multiphase refinement**
  - calculation of all dataset types can be done for multiple phases*
- **RMCProfile7 can fit**
  - real space dataset (X-Ray and neutron) can be calculated as a back Fourier Transform of reciprocal space dataset
  - full GSAS-II compatibility (easy Bragg data extraction)
- **Variety of additional constraints**
  - molecular potentials (distances, angles, torsion angle, inversion angle, planarity + variants)
  - potentials and swaps are now compatible
- **Molecule (rigid body) move type**
  - molecule (rigid body) type move
  - swap between atoms and atoms, atoms to molecules and molecules to molecules

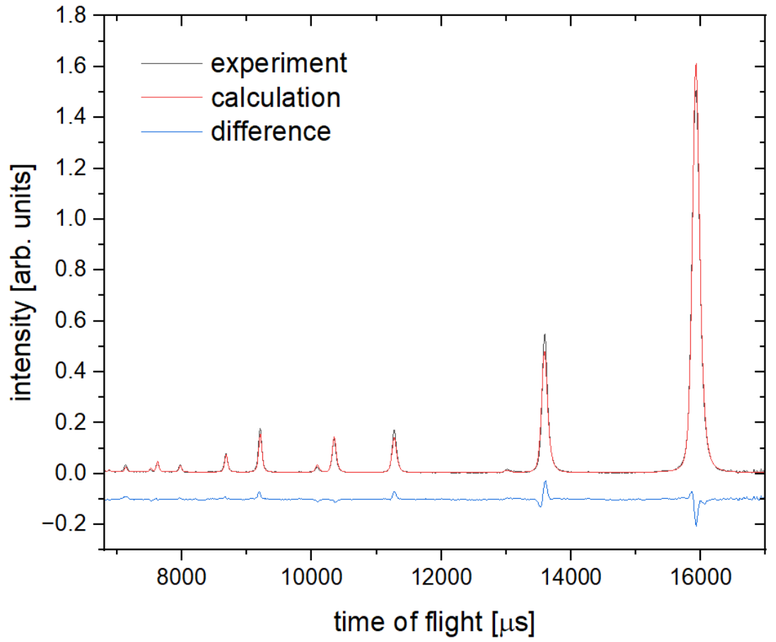
*W. Sławiński, J. Appl. Cryst. (2018). **51**, 919–923

Example 1.
$Sc_xV_{1-x}O_{2-\delta}$ X-Ray and neutron complementarity

x-ray diffraction

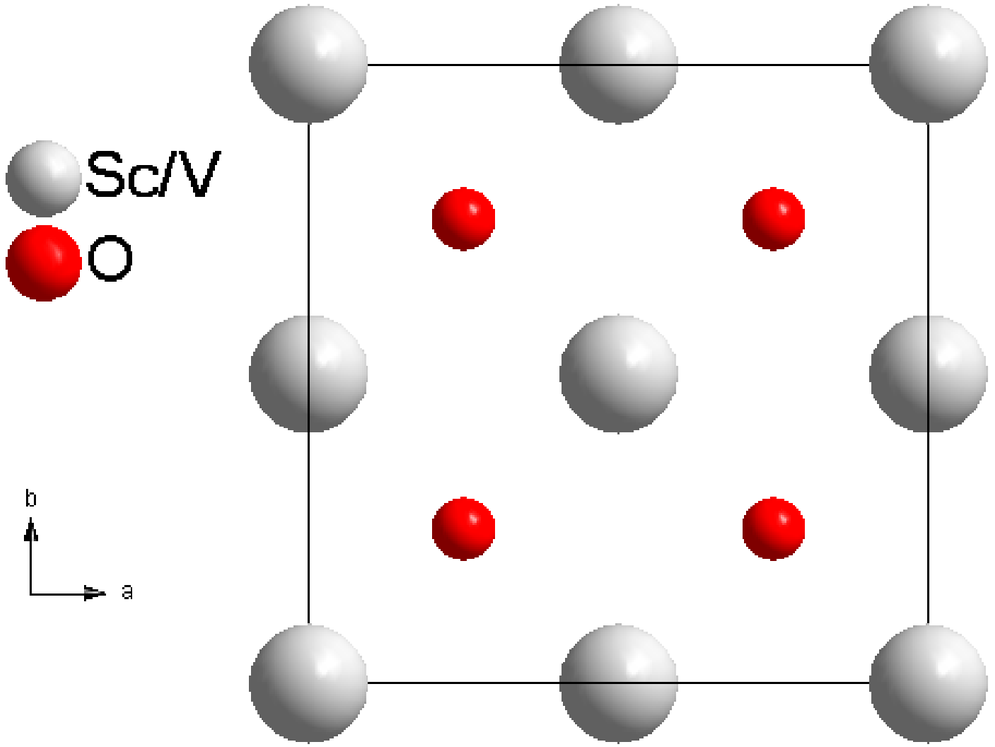


neutron diffraction



$Sc_xV_{1-x}O_{2-\delta}$ X-Ray and neutron complementarity

Average structure with δ = 0.25



General	
Origin	ScVO2.5

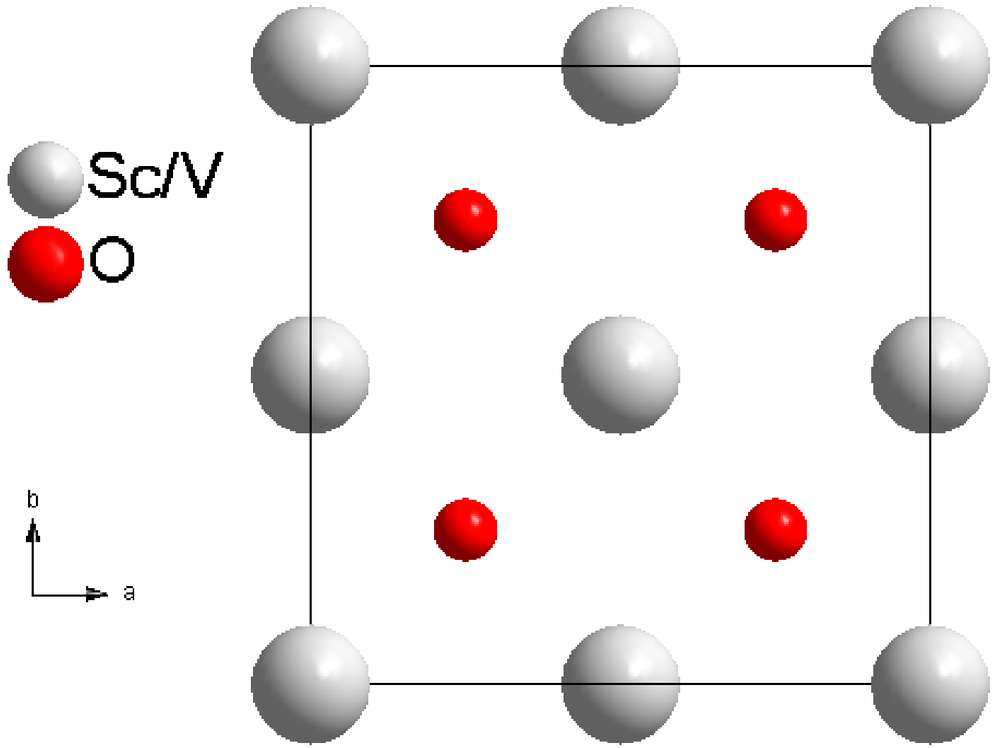
Bibliographic data

Phase data	
Space-group	F m -3 m (225) - cubic
Cell	a=4.9883 Å V=124.13 Å^3 Z=1

Atomic parameters							
Atom	Wyck.	Site	S.O.F.	x/a	y/b	z/c	U [Å^2]
V	4a	m-3m	0.3333	0	0	0	0.0620
Sc	4a	m-3m	0.6667	0	0	0	0.0435
O1	8c	-43m	0.875	1/4	1/4	1/4	0.1003

$Sc_xV_{1-x}O_{2-\delta}$ X-Ray and neutron complementarity

Average structure



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$Sc_xV_{1-x}O_{2-\delta}$ X-Ray and neutron complementarity

from **Bragg diffraction**

- unit cell size
- crystallographic lattice
- average atom positions and average distances

$$d_{ij} = \langle r_i \rangle - \langle r_j \rangle$$

- structure composition

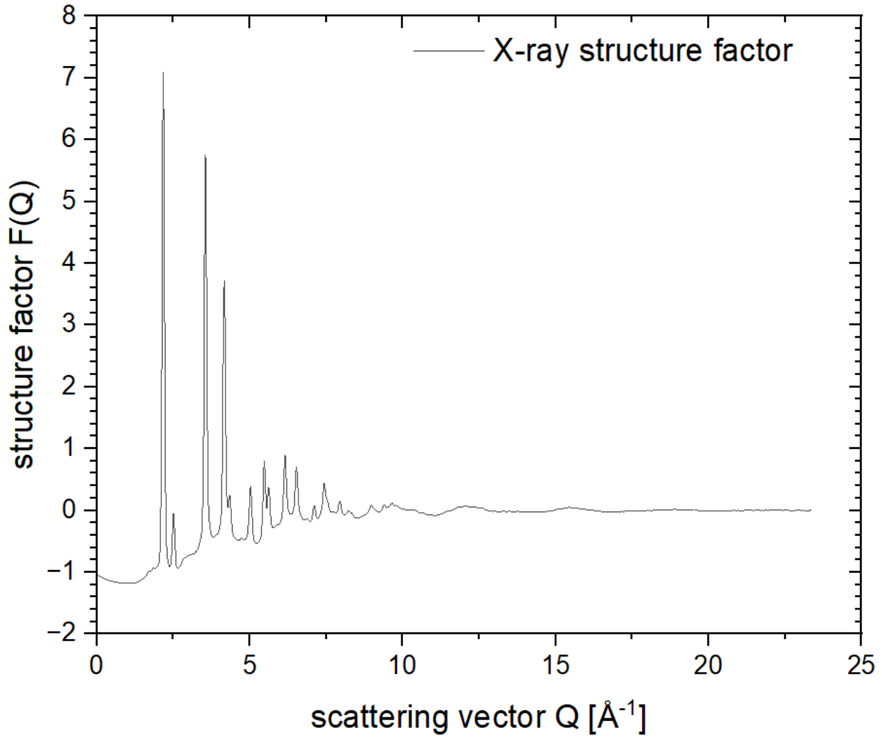
from **Total Scattering**

- local crystal structure
- local distortions and dislocations
- individual atom positions and distances

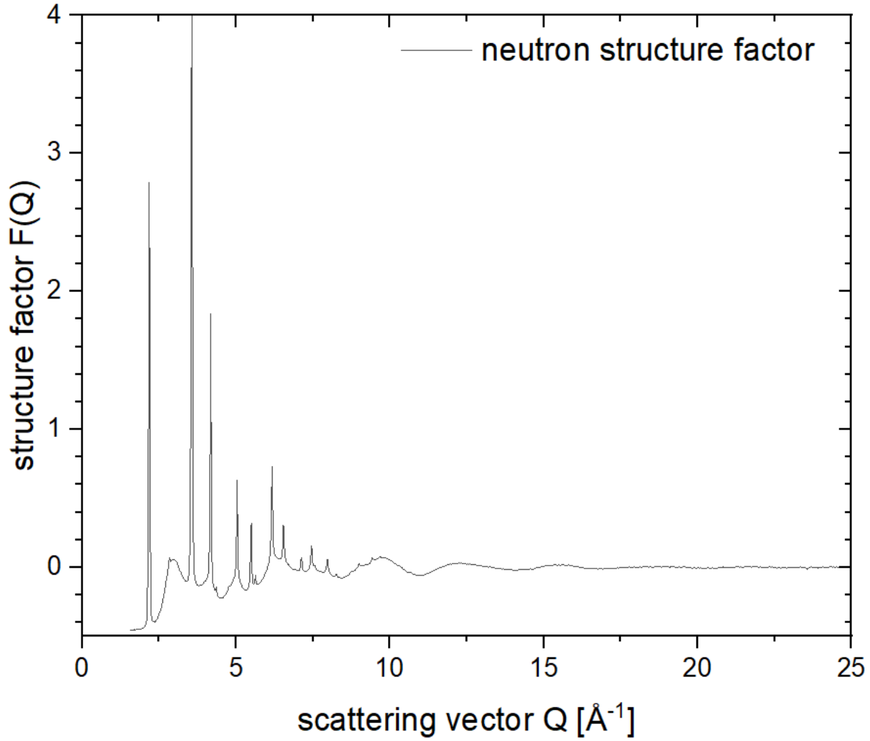
$$d_{ij} = \langle r_i - r_j \rangle$$

$Sc_xV_{1-x}O_{2-\delta}$ X-Ray and neutron complementarity

x-ray diffraction



neutron diffraction



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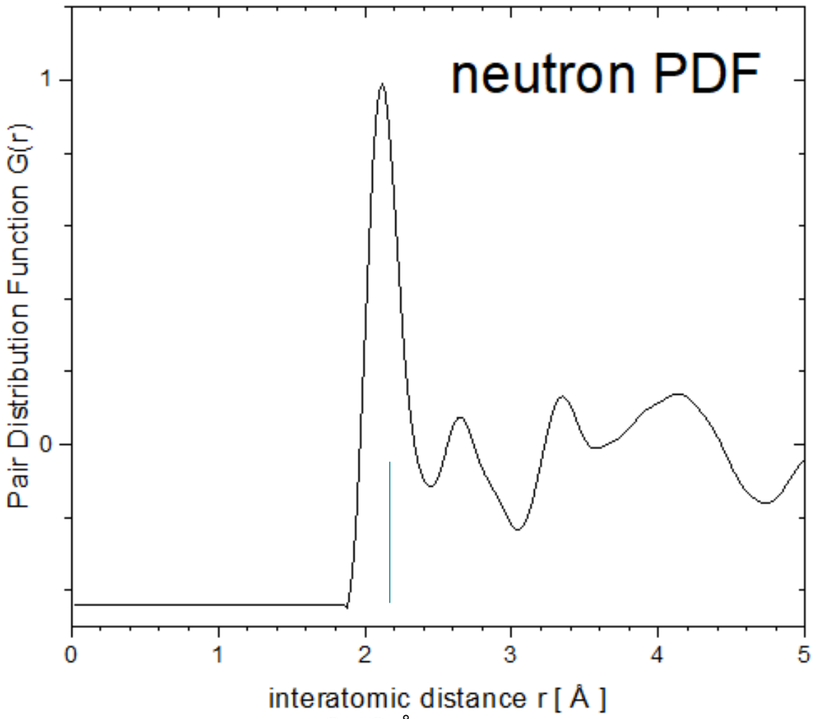
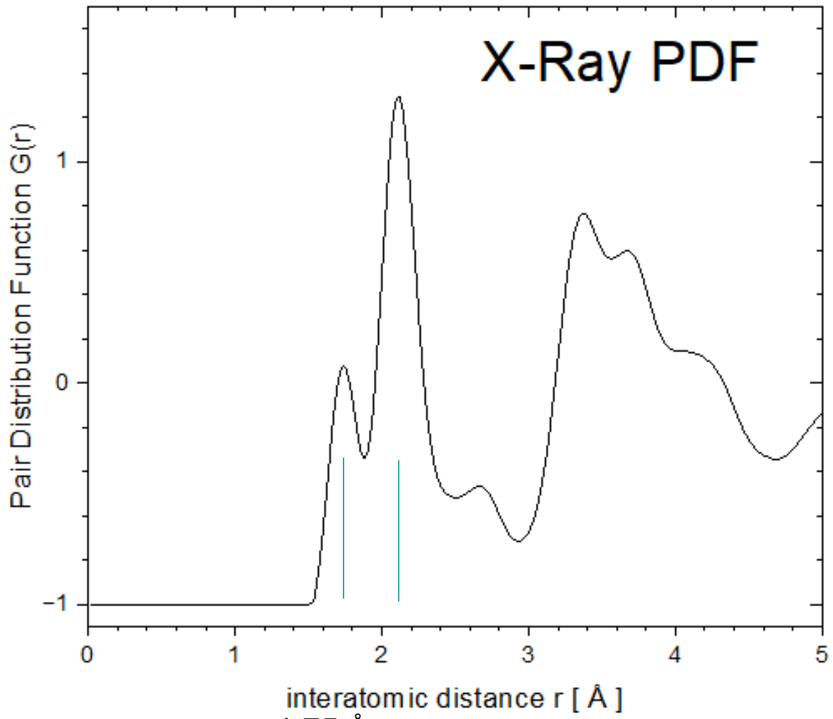
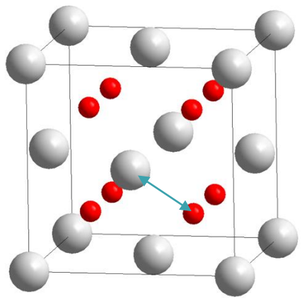
$Sc_xV_{1-x}O_{2-\delta}$ X-Ray and neutron complementarity







$d_{Sc/V-O}$ = 2.16 Å (from average structure)

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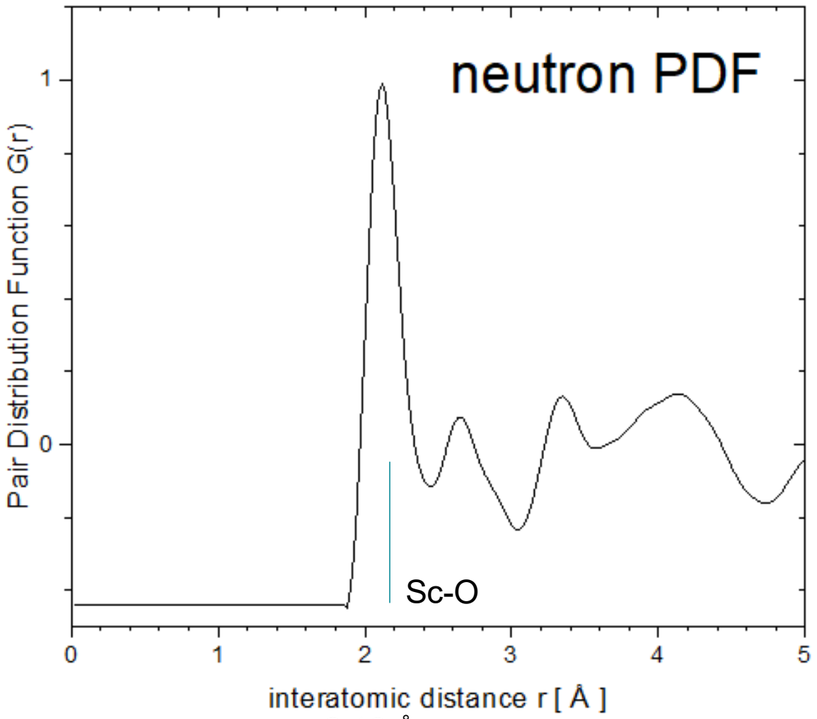
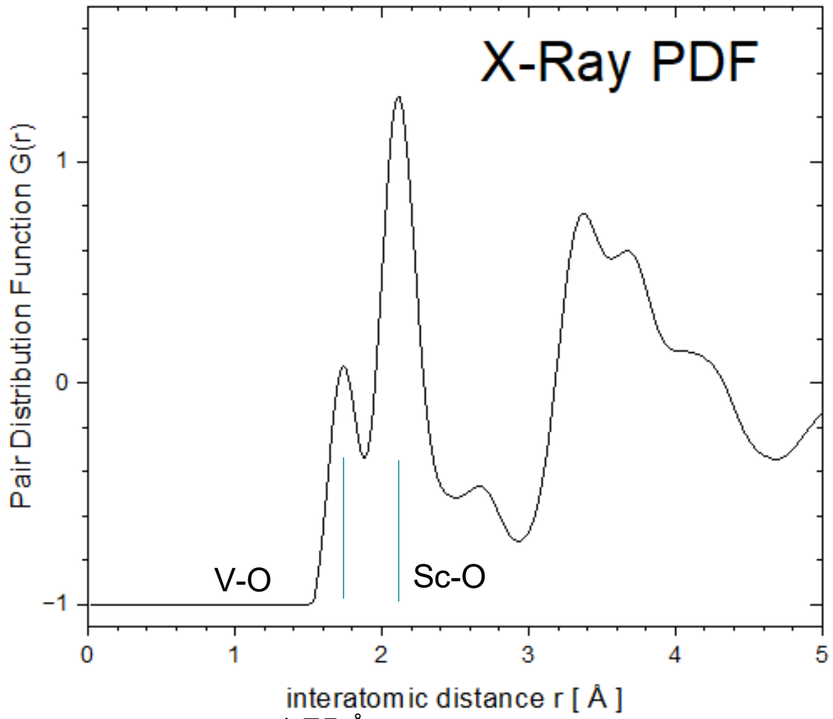
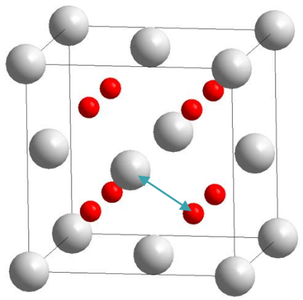
$Sc_xV_{1-x}O_{2-\delta}$ X-Ray and neutron complementarity







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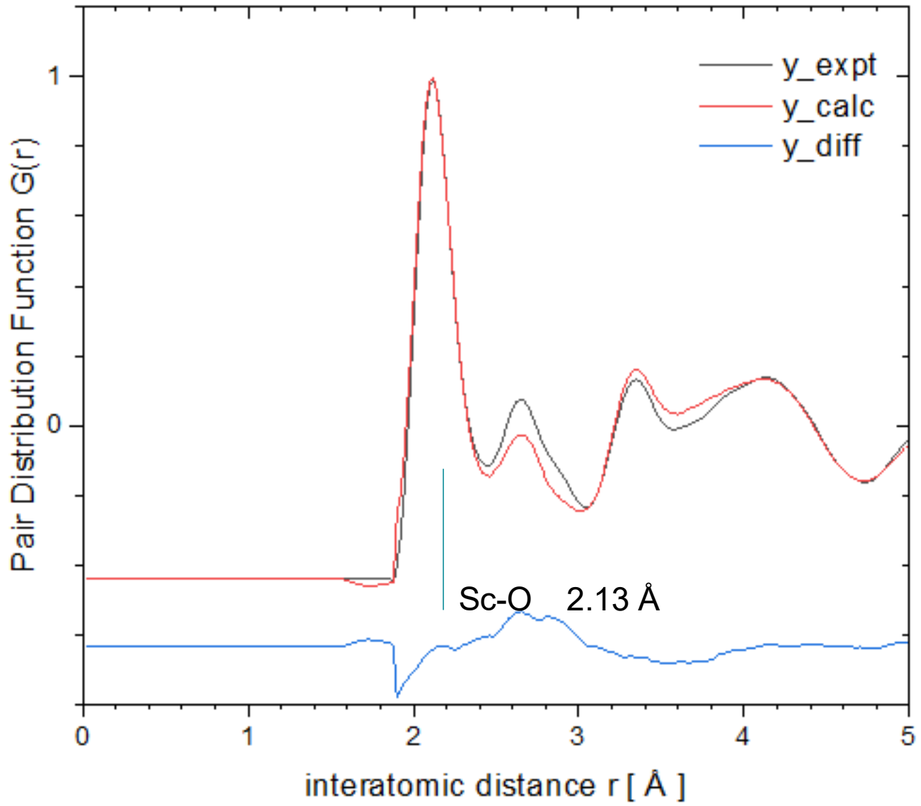
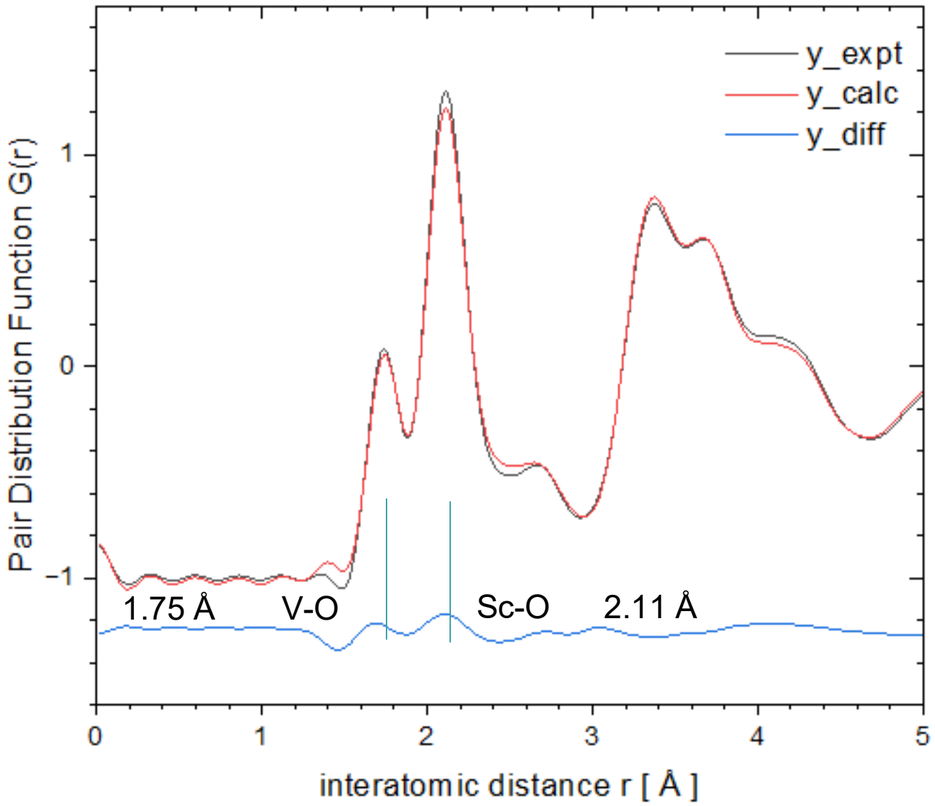
$Sc_xV_{1-x}O_{2-\delta}$ X-Ray and neutron complementarity





neutrons	Sc	V	O
Sc	0.202	-0.006	0.501
V		0.000	-0.008
O			0.311

X-Rays	Sc	V	O
Sc	0.154	0.169	0.308
V		0.046	0.169
O			0.154

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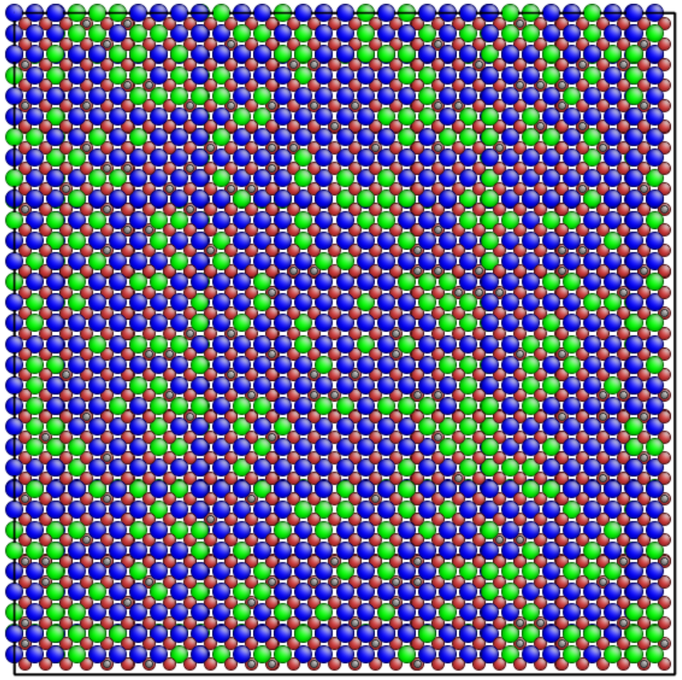
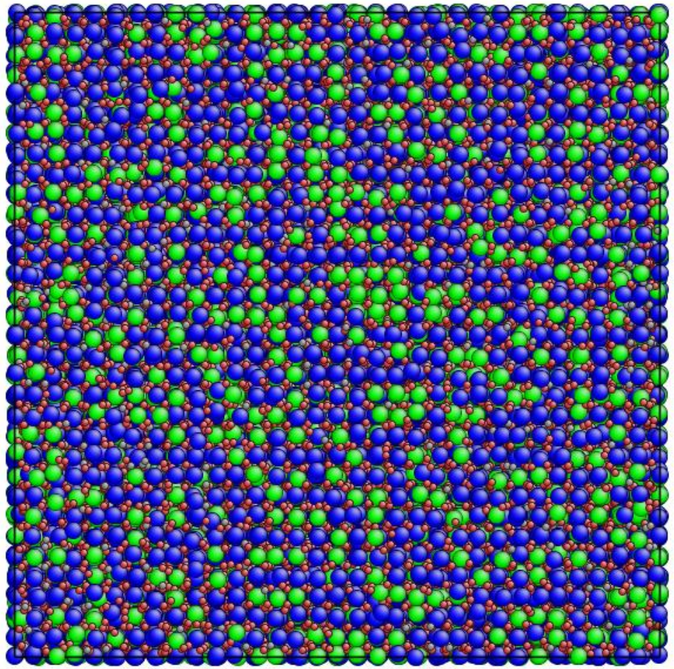
$Sc_xV_{1-x}O_{2-\delta}$ initial vs final configuration

initial configuration

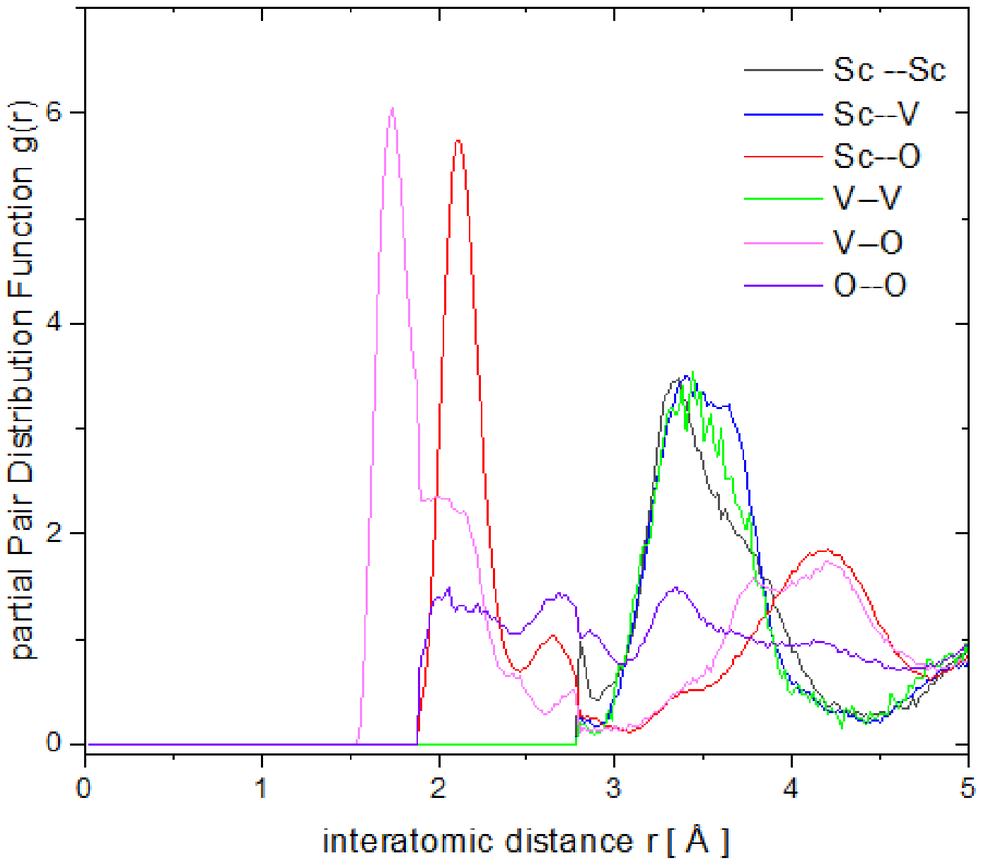


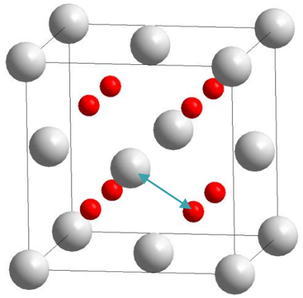
16x16x16 supercell

final configuration



$Sc_xV_{1-x}O_{2-\delta}$ parials





$d_{Sc/V-O}$ = 2.16 Å (from average structure)

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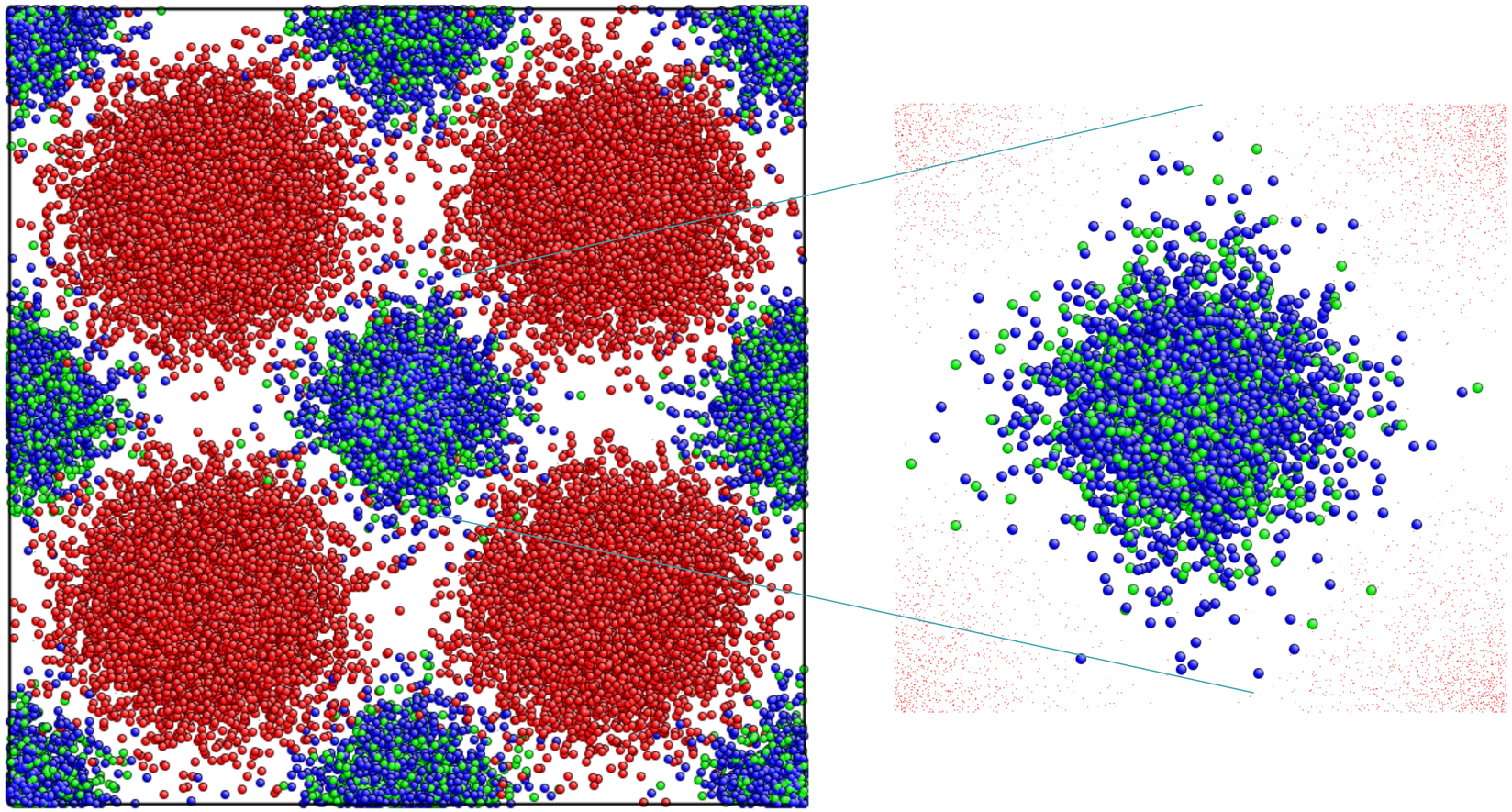
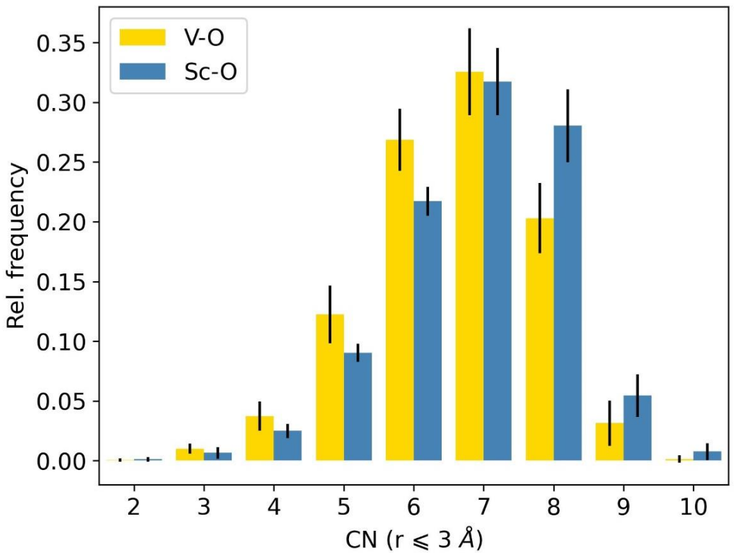
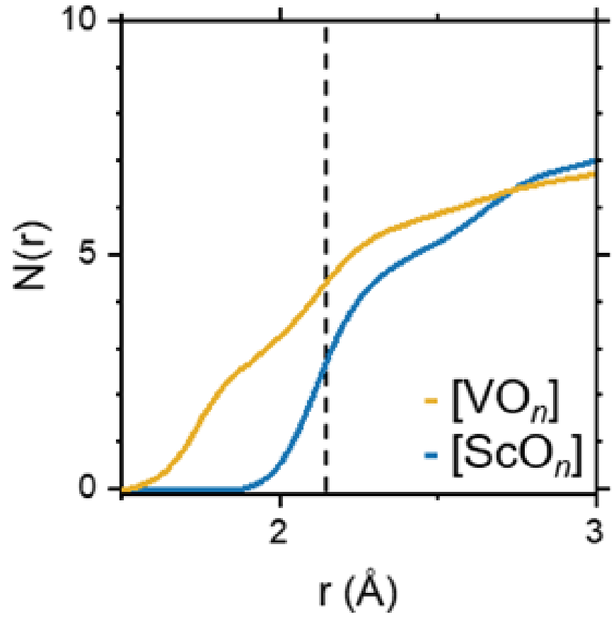
$Sc_xV_{1-x}O_{2-\delta}$ post refinement analysis



$Sc_xV_{1-x}O_{2-\delta}$ post refinement analysis



Distribution of coordination numbers for $[VO_n]$ (yellow) and $[ScO_n]$ (blue) polyhedra in c-$Sc_2VO_{5.25}$ obtained from 10 independent RMC refinements. Bond lengths are limited to r_{max} = 3.0 Å.



Coordination numbers for $[VO_n]$ (yellow) and $[ScO_n]$ (blue) polyhedra in c-$Sc_2VO_{5.25}$ Bond lengths are limited to r_{max} = 3.0 Å.

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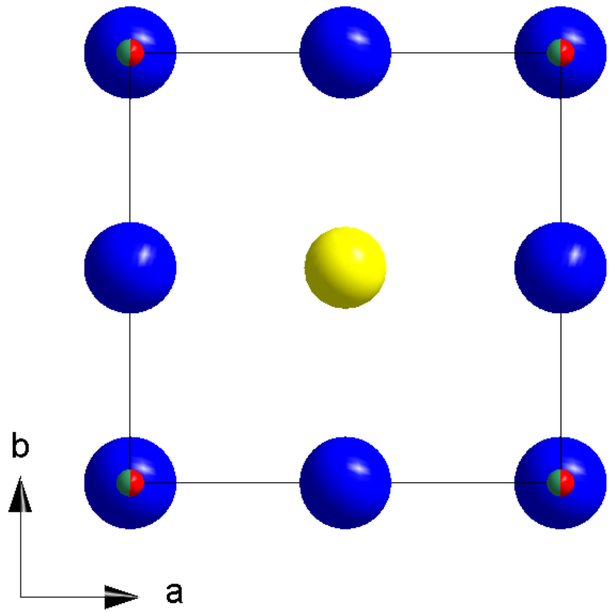
Example 2.
$BaSc_xTi_{1-x}O_{3-\delta}$ neutron elemental contrast

$BaSc_{0.5}Ti_{0.5}O_{3-\delta}$

neutron diffraction

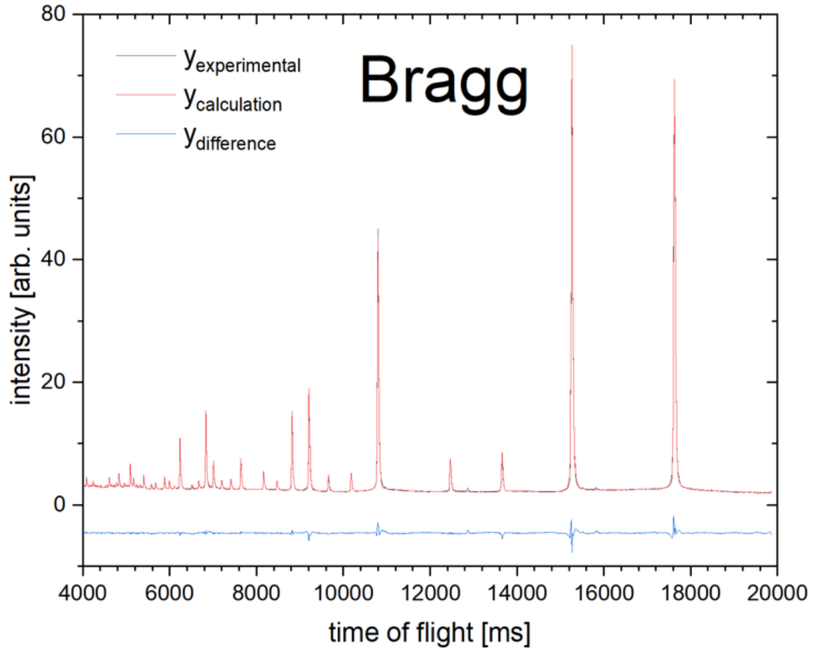


General	
Origin	BASCTIO3_0_50_0

Bibliographic data

Phase data	
Space-group	P m -3 m (221) - cubic
Cell	a=4.1306(1) Å V=70.47(1) Å^3 Z=1

Atomic parameters						
Atom	Wyck.	Site	S.O.F.	x/a	y/b	z/c
BA1	1b	m-3m		1/2	1/2	1/2
SC2	1a	m-3m	0.491	0	0	0
TI3	1a	m-3m	0.509	0	0	0
O4	3d	4/mm.m	0.92267	1/2	0	0



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Example 2.
$BaSc_xTi_{1-x}O_{3-\delta}$ neutron elemental contrast

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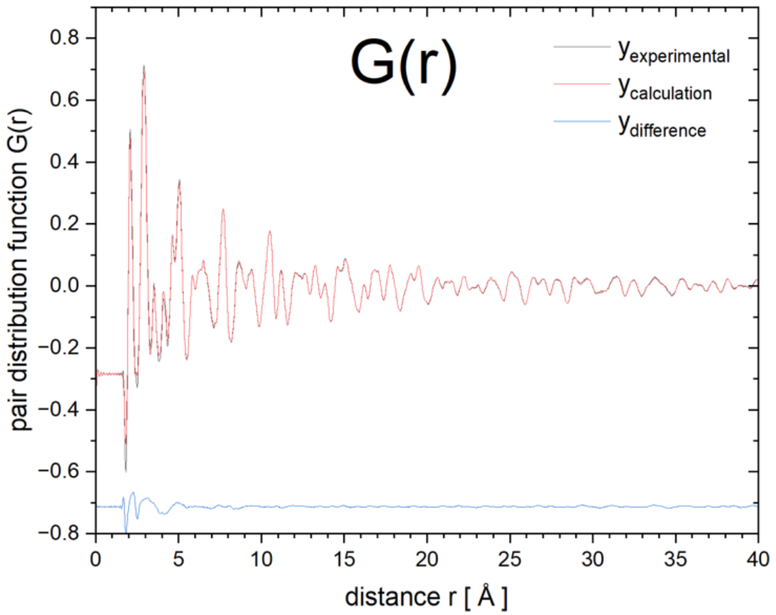
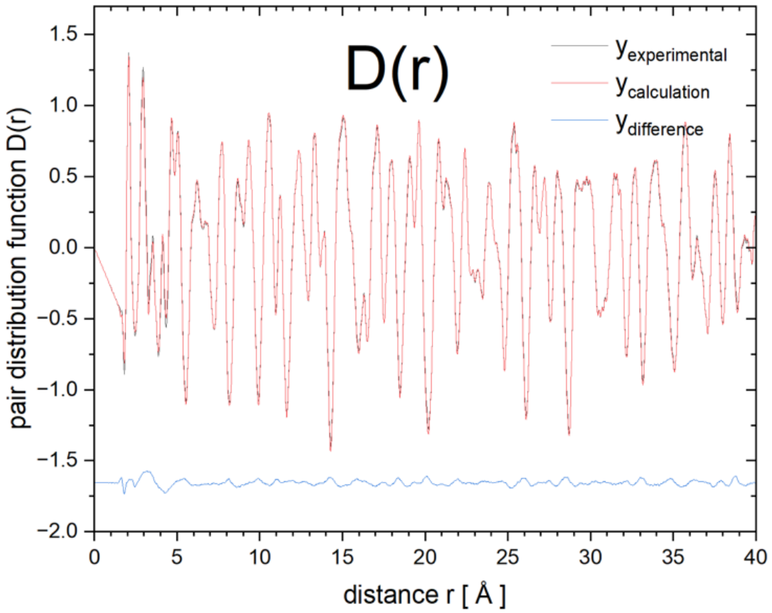
neutron diffraction





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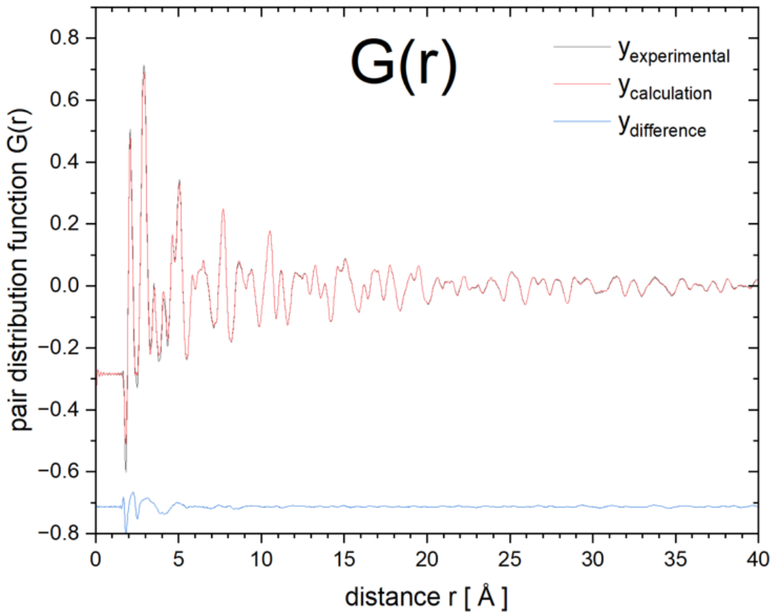
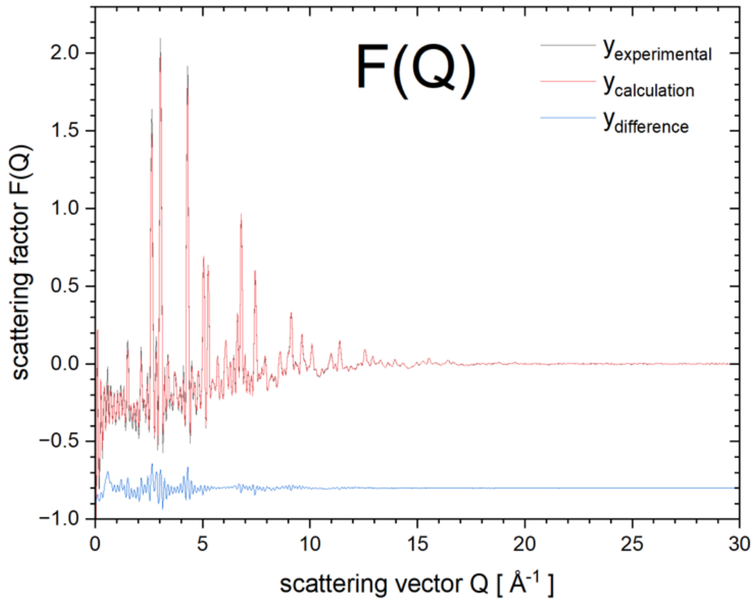
$BaSc_{0.5}Ti_{0.5}O_{3-\delta}$

neutron diffraction





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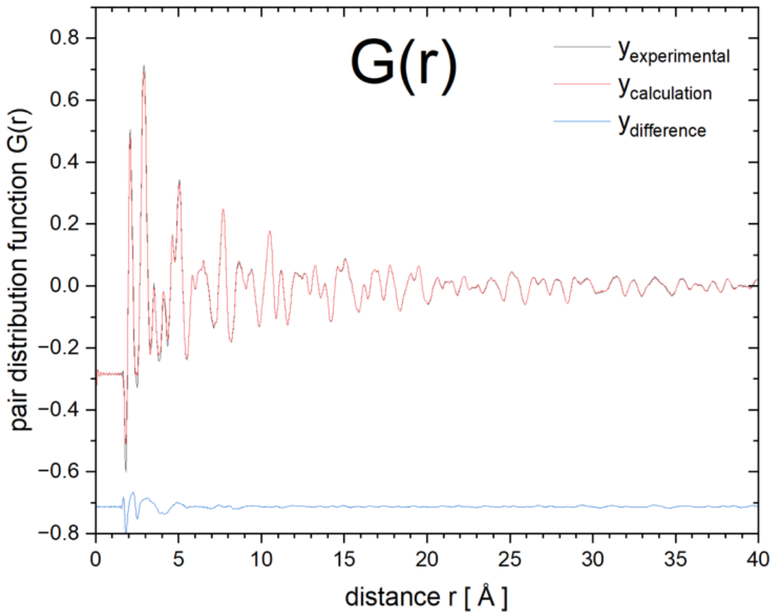
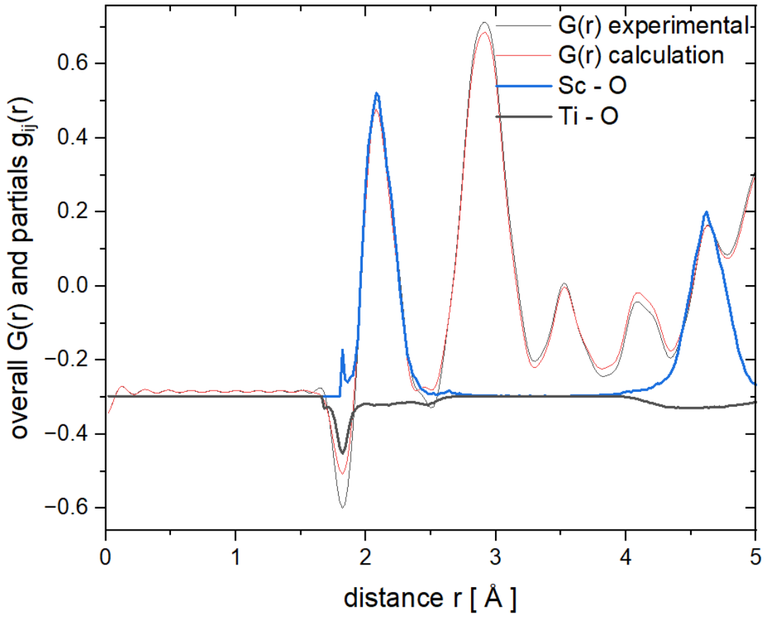
Example 2.
$BaSc_xTi_{1-x}O_{3-\delta}$ neutron elemental contrast

$BaSc_{0.5}Ti_{0.5}O_{3-\delta}$

neutron diffraction





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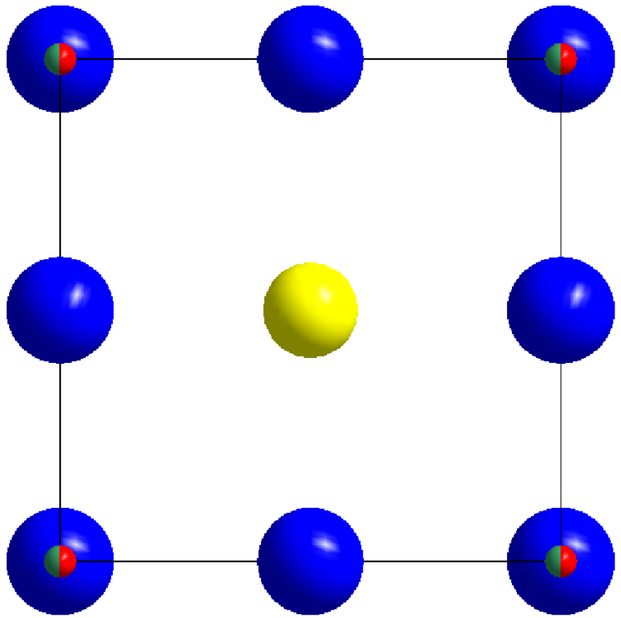
Example 2.
$BaSc_xTi_{1-x}O_{3-\delta}$ neutron elemental contrast

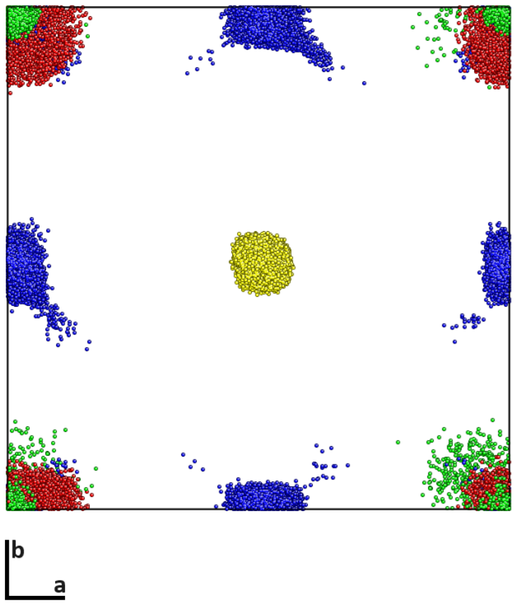
$BaSc_{0.5}Ti_{0.5}O_{3-\delta}$

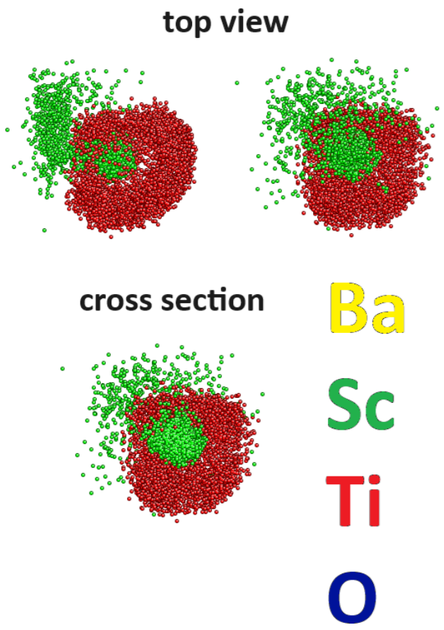
neutron diffraction

average

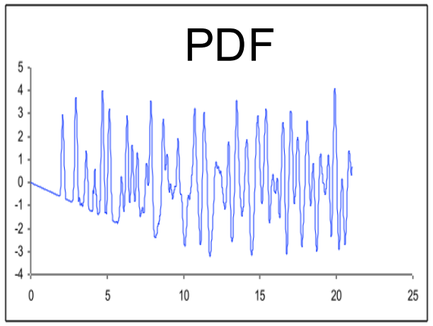


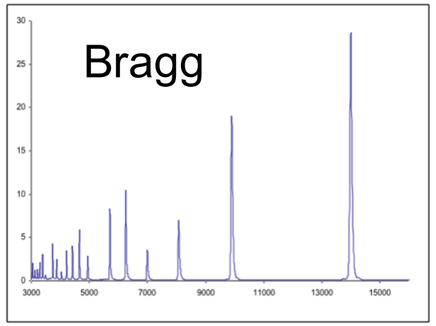
big box local structure modelling

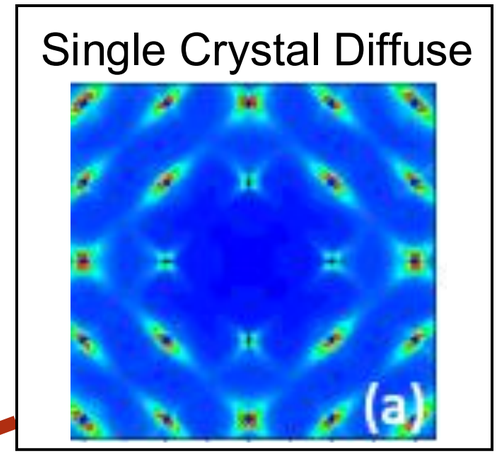


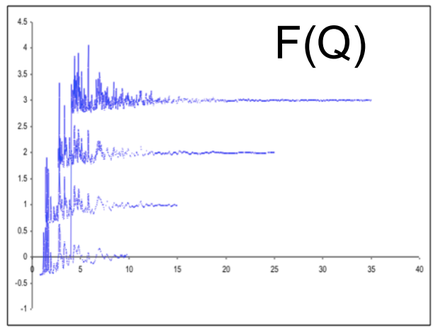


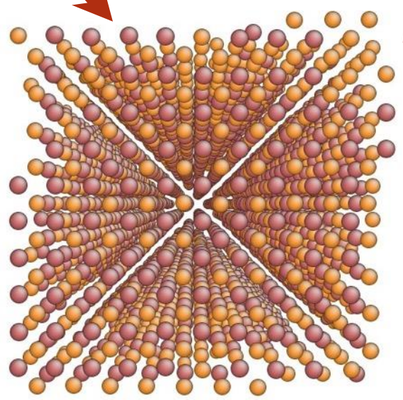
RMC PROFILE

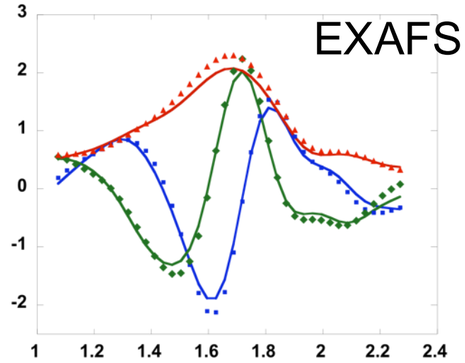


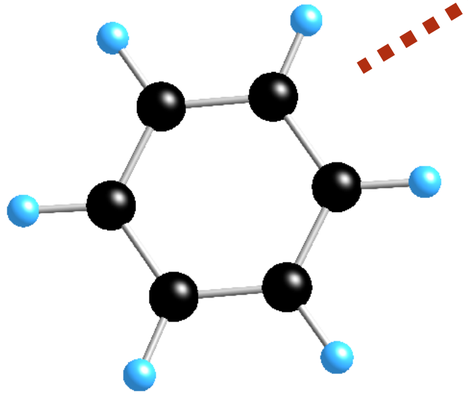




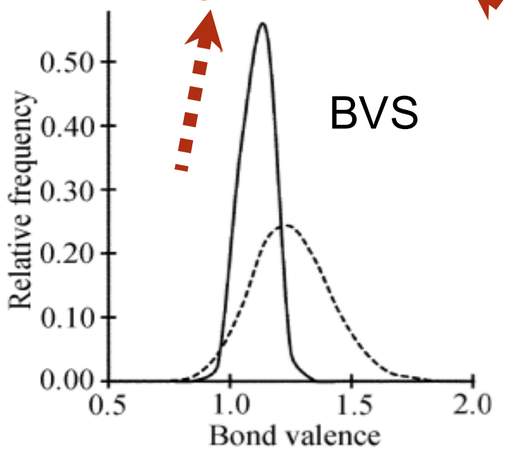


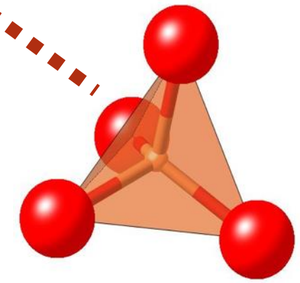






Molecular potentials





Polyhedral restraints

M G Tucker et al, *J. Phys.: Condens. Matter* **19,** 335218 (2007).

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Acknowledgements

Martin Dove (QMUL)

Dave Keen (ISIS)

Andrew Goodwin (Oxford)

Stefan Norberg (Chalmers)

Igor Levin (NIST)

Victor Krayzman (NIST)

Helen Playford (ISIS)

Wojciech Slawinski (Warsaw)

Marshall McDonnell (ORNL)

Yuanpeng Zhang (ORNL)

Maksim Eremenko (ORNL)

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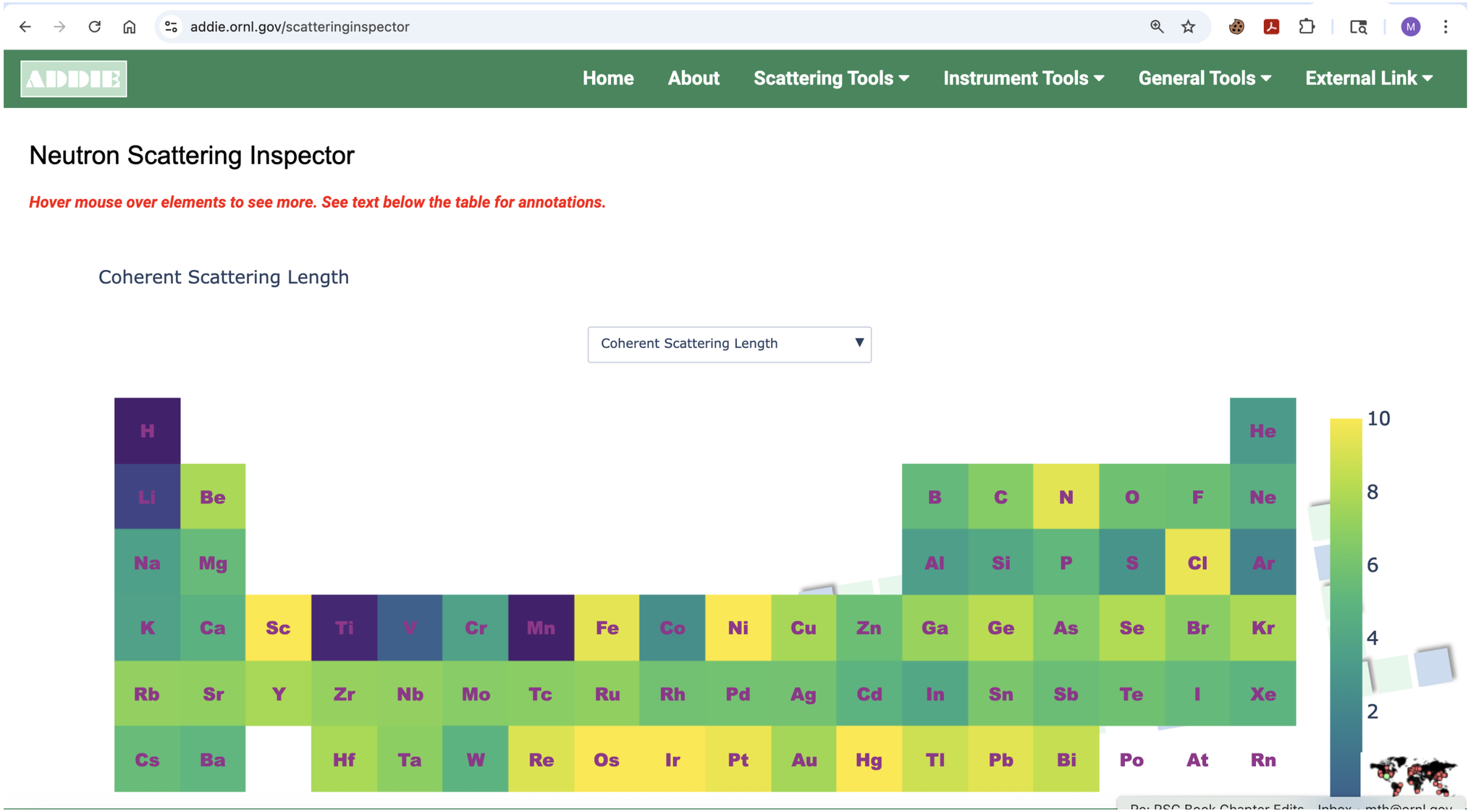

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Thank you!

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- **wslawinski@chem.uw.edu.pl**

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addie.ornl.gov