The ILL Diffraction Group

www.ill.fr/dif/

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Diffraction Group

- Largest of 5 instrument groups at ILL
- 10 permanent staff scientists (including 3 Australians !)
- Total of ~30 scientists, students and technicians
- 3 of the first 5 new “Millennium” projects
- 2 more Millennium projects in the second tranche.

C.f. New Australian reactor

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The Millennium Programme at ILL -> New Neutron Detectors

New Diffraction Group Instruments:

- D20 – a large microstrip detector for chemical kinetics…
- D4c – a microstrip detector for liquids & amorphous materials
- Strain Scanner – for mapping strain using microstrip detectors
- D19 – an array of 2D-microstrips for protein/fibre diffraction
- T-LADI – Laue Diffractometer & neutron Image plate detector
- D2b – high resolution powder diffractometer with linear PSDs
- D3c – He3 neutron spin filters and magnetic polarimetry

The Millennium Programme at ILL
-> New Neutron Detectors

Other Existing Diffraction Group Instruments:

- D1a – first high resolution powder diffractometer
- D1b – first high flux position sensitive detector (CNRS-CRG)
- D9 - first hot source, 4-circle machine (PSD, lifting detector)
- D10 – 4-circle, 3-axis diffractometer (Garry McIntyre et al.)
- D15 – 2-axis/4-circle diffractometer (CENG-CRG)
- D23 – new 2-axis polarised neutron machine (CENG-CRG)
- S42 – Laue camera for crystal alignment (Marmeggi)

The Millennium Programme at ILL
-> New Neutron Detectors

Investment in reactors & other neutron sources is necessary,

but…

Investment in *detecting* more neutrons is very cost effective

and we need…

Microstrip detectors, neutron image plates, detector arrays…

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What is a Microstrip Detector ?

Instead of wires, a printed circuit is used.

This allows high resolution, mechanical stability...

Microstrip Detectors - Printed Circuits

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"Mr Microstrip" Anton Oed with admirer
(Giovanna Cicognani, ILL Science Secretary)

ILL Detector Group:

- Bruno Guerard (head)
- Jean-Francois Clergeau
- Dominique Feltin
- Michel Gamon
- Giuliana Manzin
- Alexandre Sicard
- Fabrice Horst
- Anton Oed (retired)

The 160º D20 Microstrip Array

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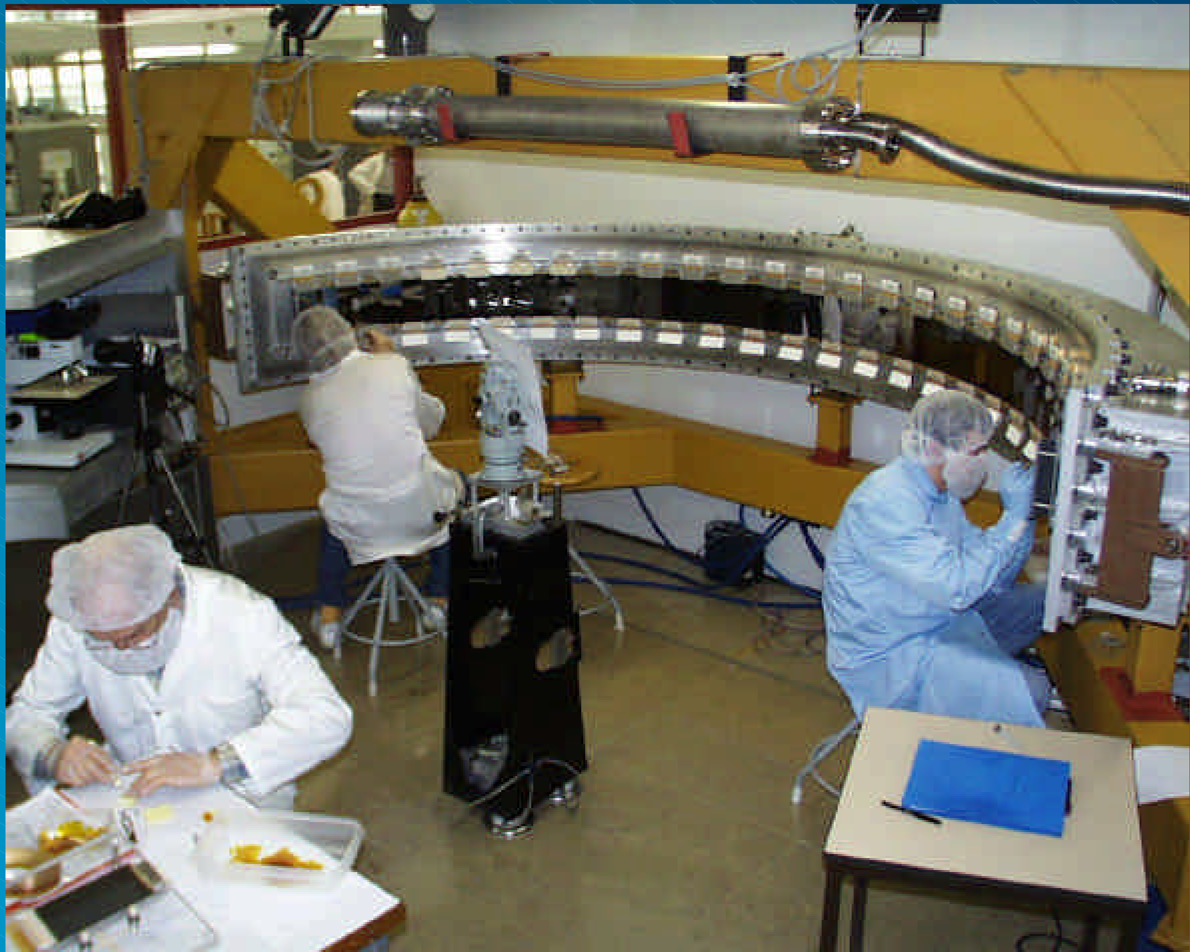

25 plates of 64 electrodes are assembled to produce a 1600-wire detector covering 160º.

High Flux Powder Diffractometer D20

Pierre Convert, Thomas Hansen, Jacques Torregrossa

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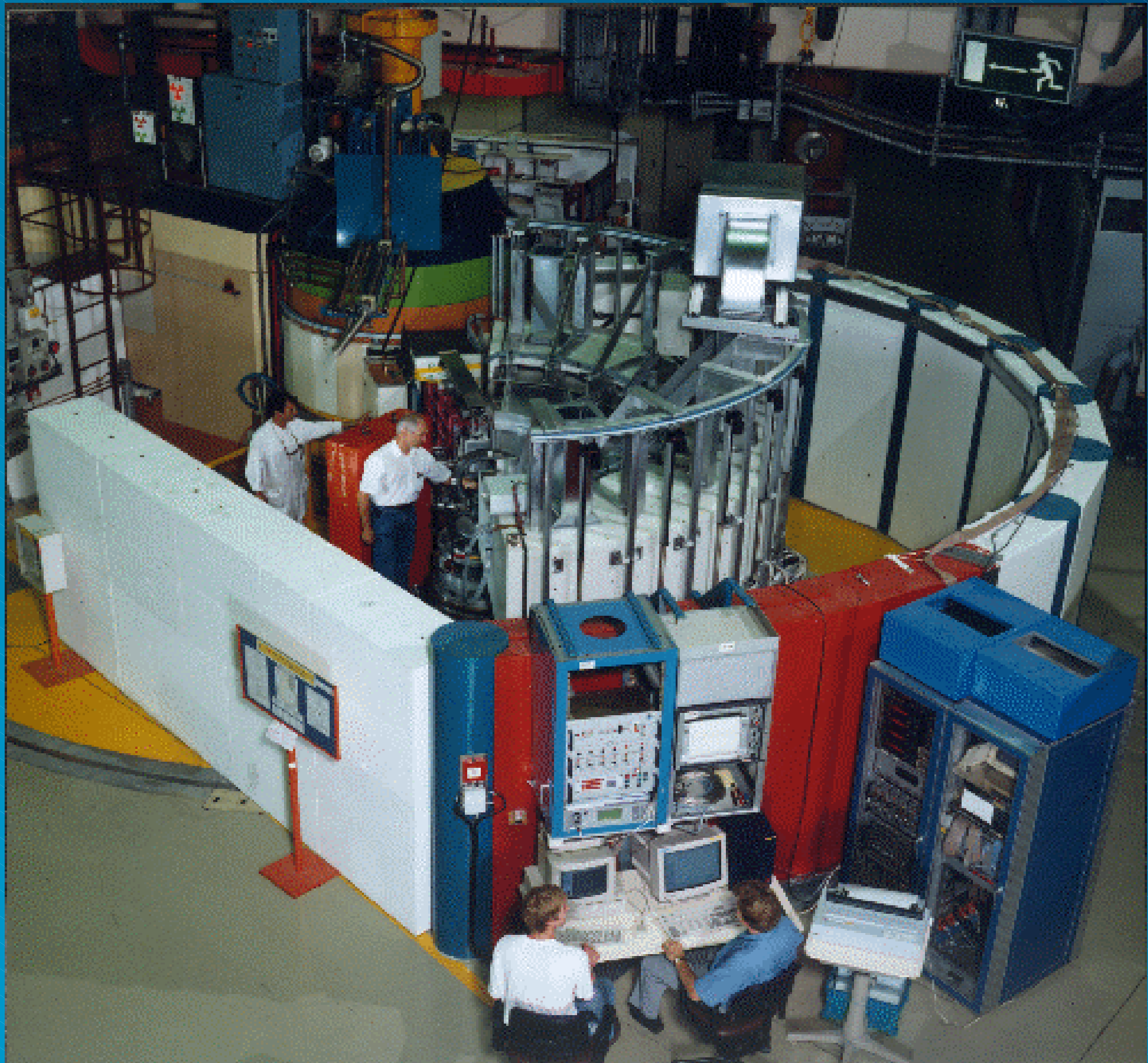

D20 in action with
Jacques Torregrossa,
Pierre Convert
& Thomas Hansen

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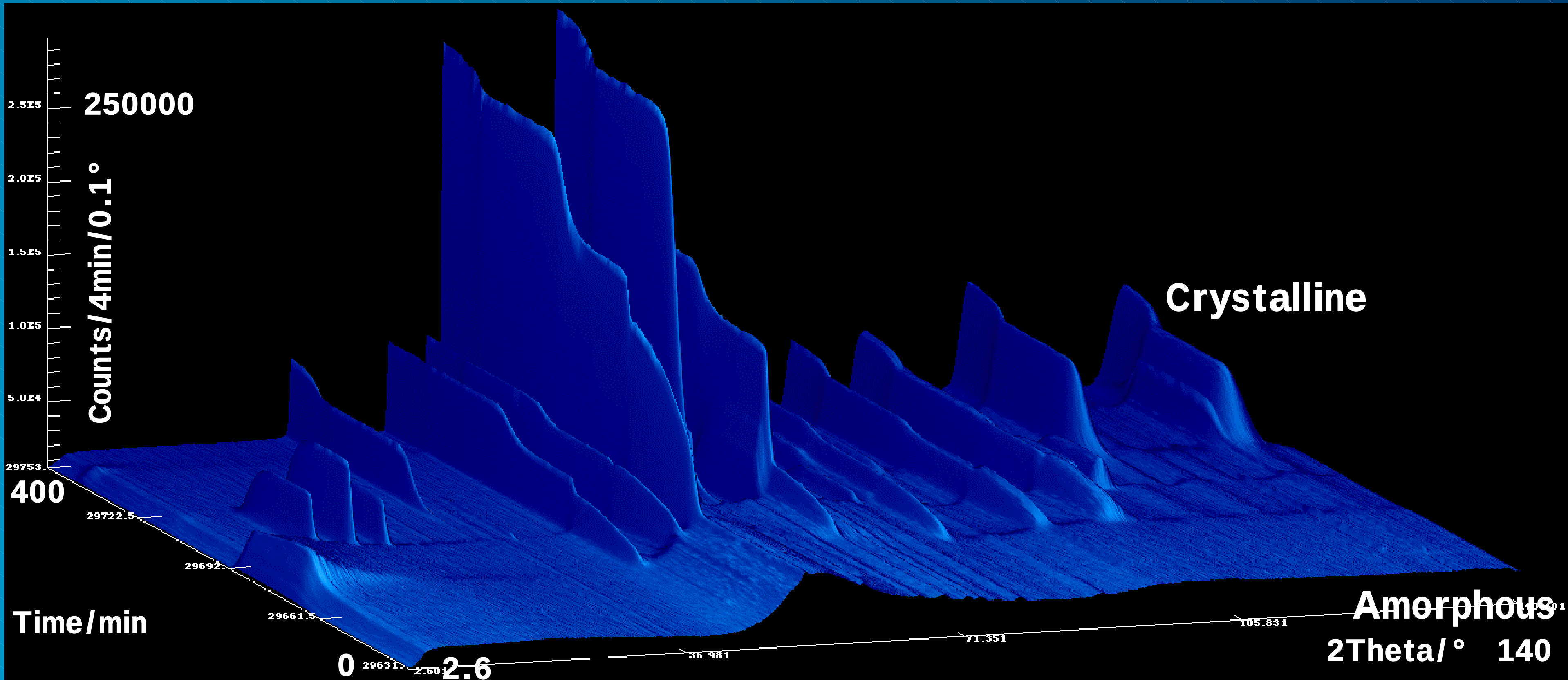
Applications of large fast detectors
Real-time Phase Diagrams

Sue Kilcoyne, Bob Cywinski et al.

Crystallisation of amorphous alloys $Y_{67}Fe_{33}$ with increasing temperature



Complete diffraction pattern in minutes or seconds, scan through temperature

Applications of large fast detectors

Real-world samples

- Neutrons much more penetrating than X-rays
  - Can use "real-world samples"
  - Eg a real battery
- NB Very short λ X-rays from Synchrotron sources are also very penetrating, BUT
  - Impose very low angle scattering

Applications of large fast detectors

Real-time electro-chemistry

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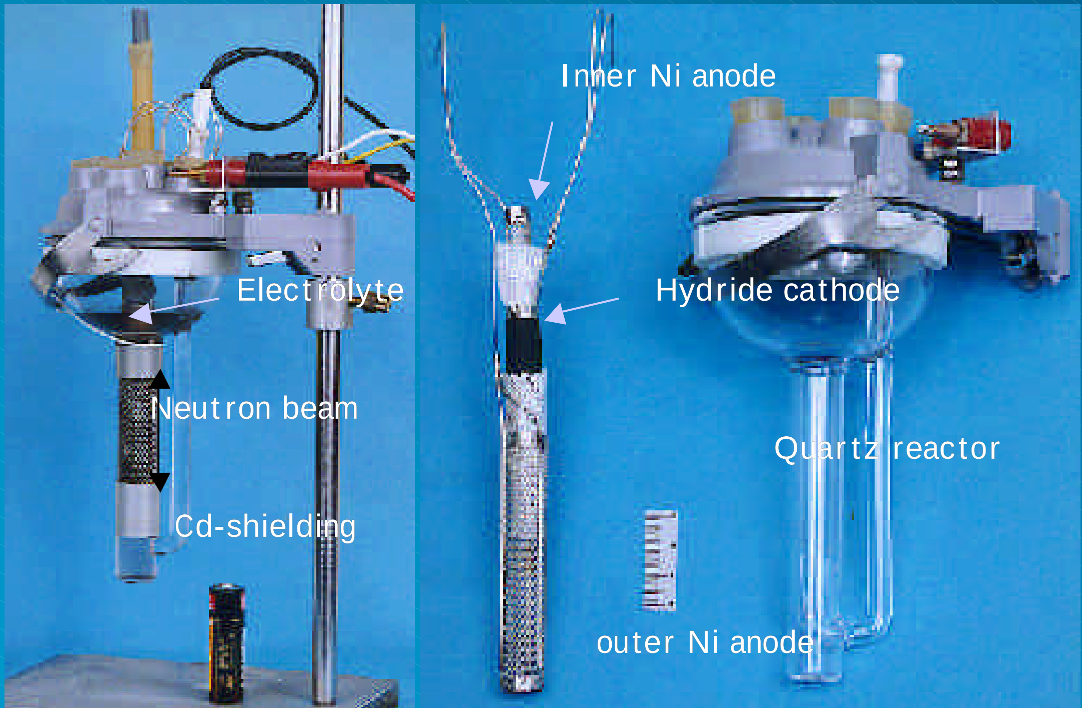
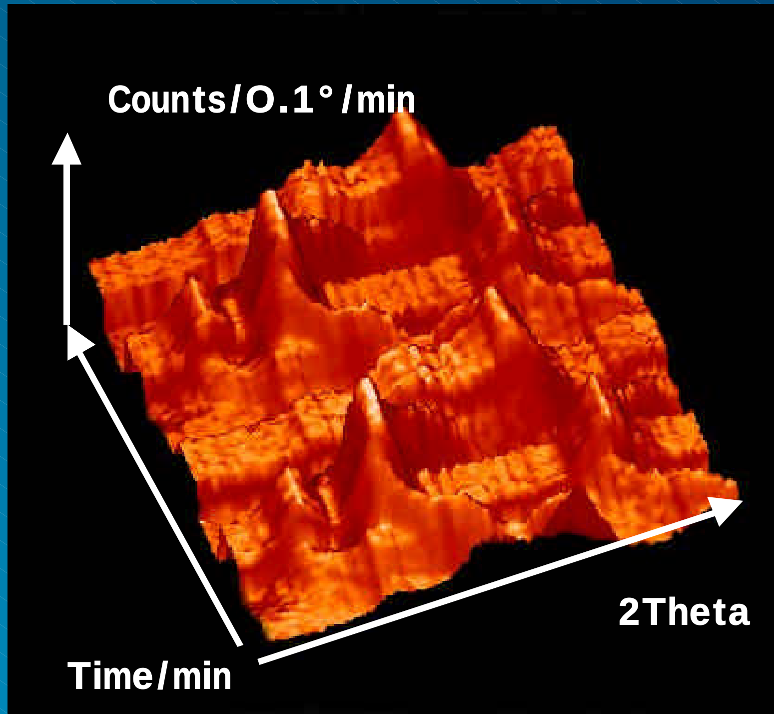
Latroche, Chabre et al.:

In-situ Charging and discharging of metal hydride electrodes LaNi5





- Follow chemical changes with battery charge/discharge cycle

High Pressure Microstrip Detectors

New D4C Liquids & Amorphous Materials Diffractometer

Henry Fischer, Gabriel Cuello, Pierre Palleau

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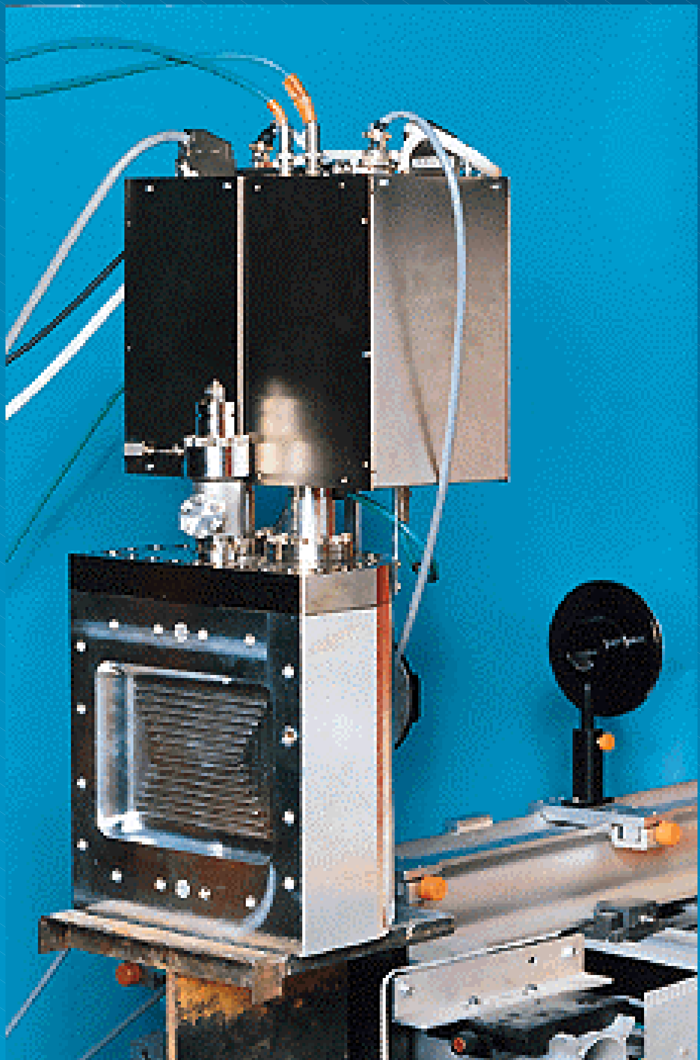
Diffraction Group



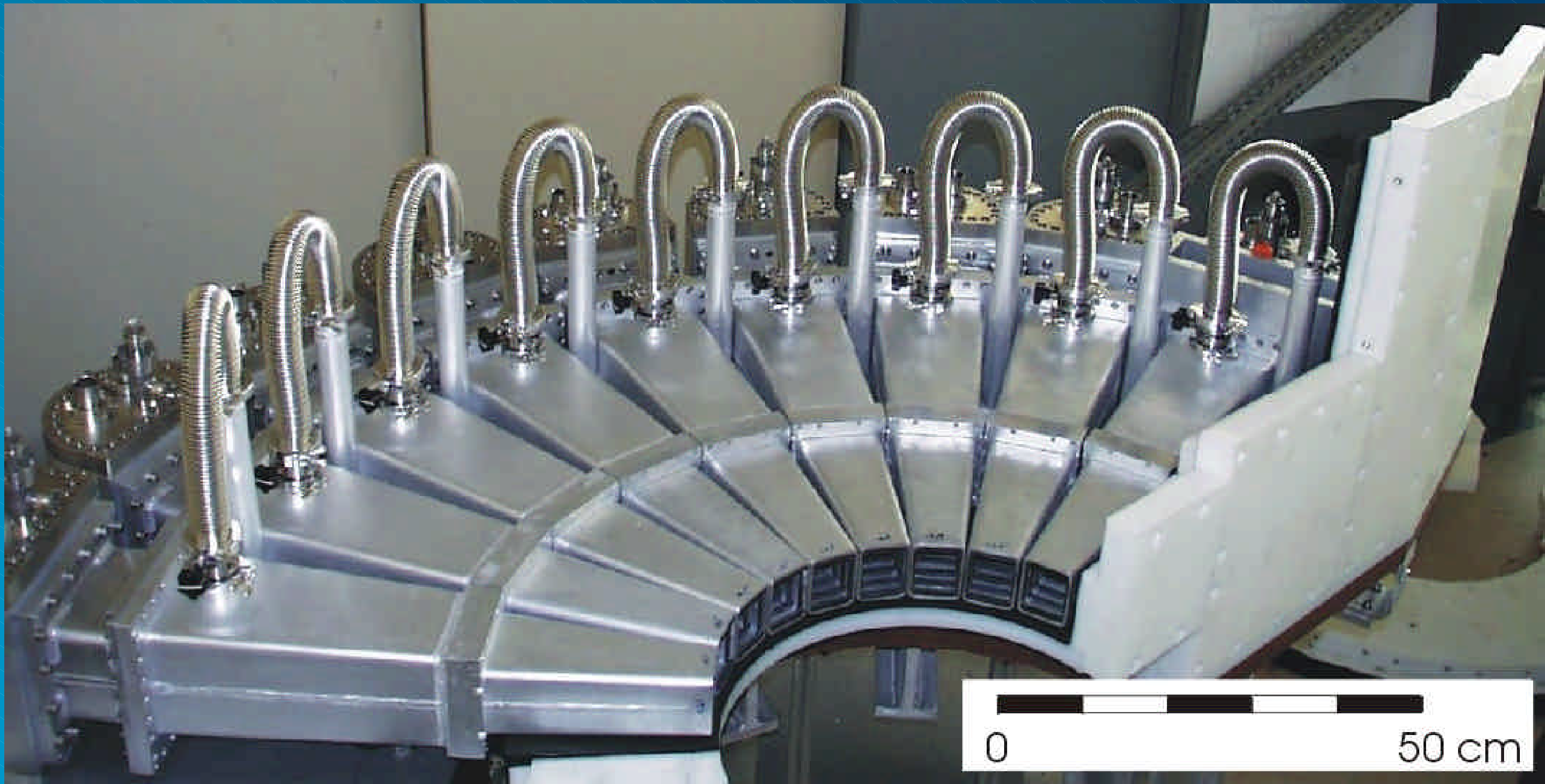
High pressure (15 bar) is needed for high efficiency at the short wavelengths needed for liquids diffraction.

The prototype D4C detector

An array of Microstrip Detectors

New D4C Liquids & Amorphous Materials Diffractometer

Henry Fischer, Gabriel Cuello, Pierre Palleau

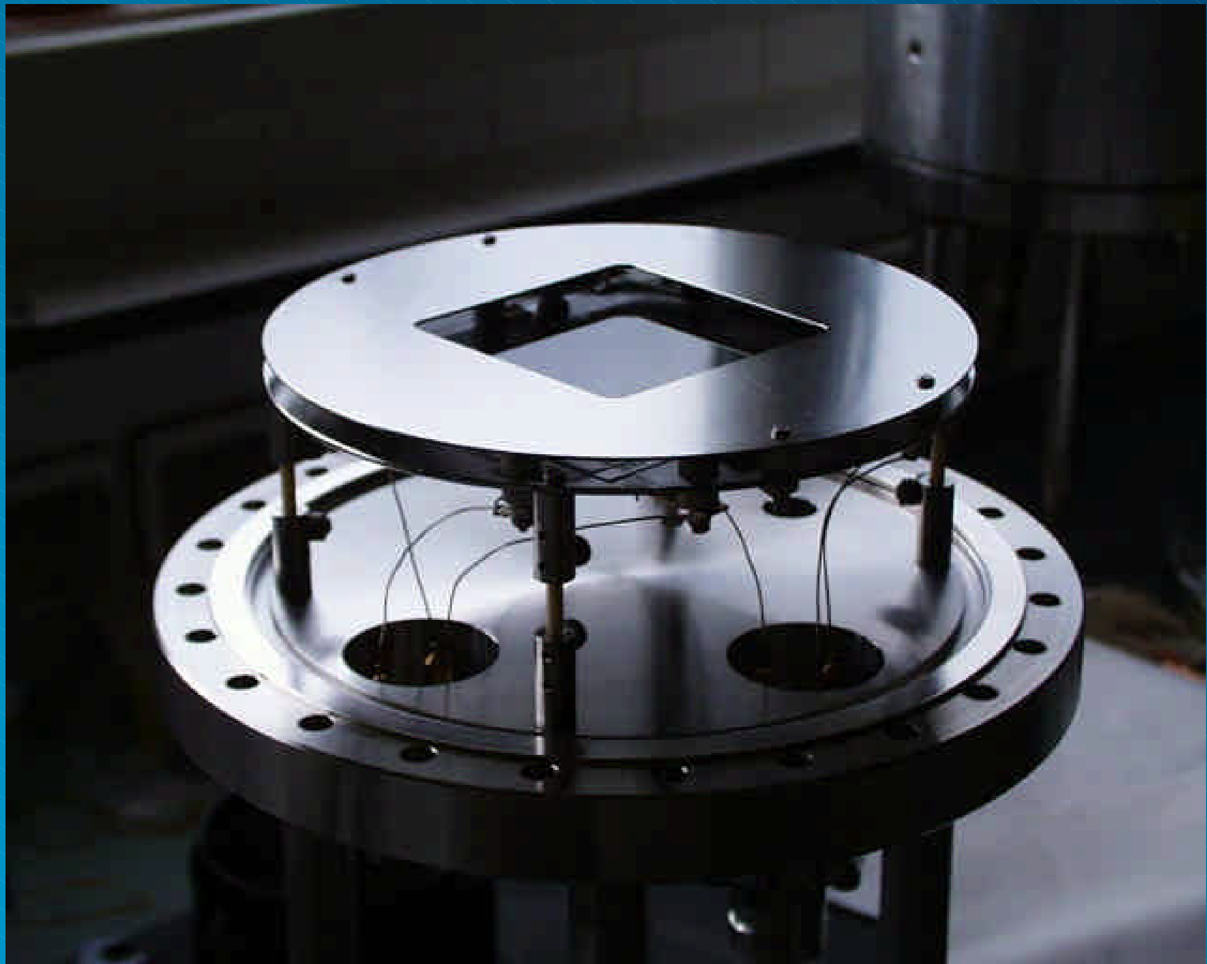


Very high efficiency & stability needed for isotope replacement method

A 2D Microstrip Detector

D9, D10, D15, Neutron Strain Scanner…

Bruno Guerard, Anton Oed et al.



A printed circuit on BOTH sides of the glass substrate

Neutron Strain Scanner

80x80 mm 2D Microstrip Detector on D1A

Thilo Pirling, Robert Wimpory

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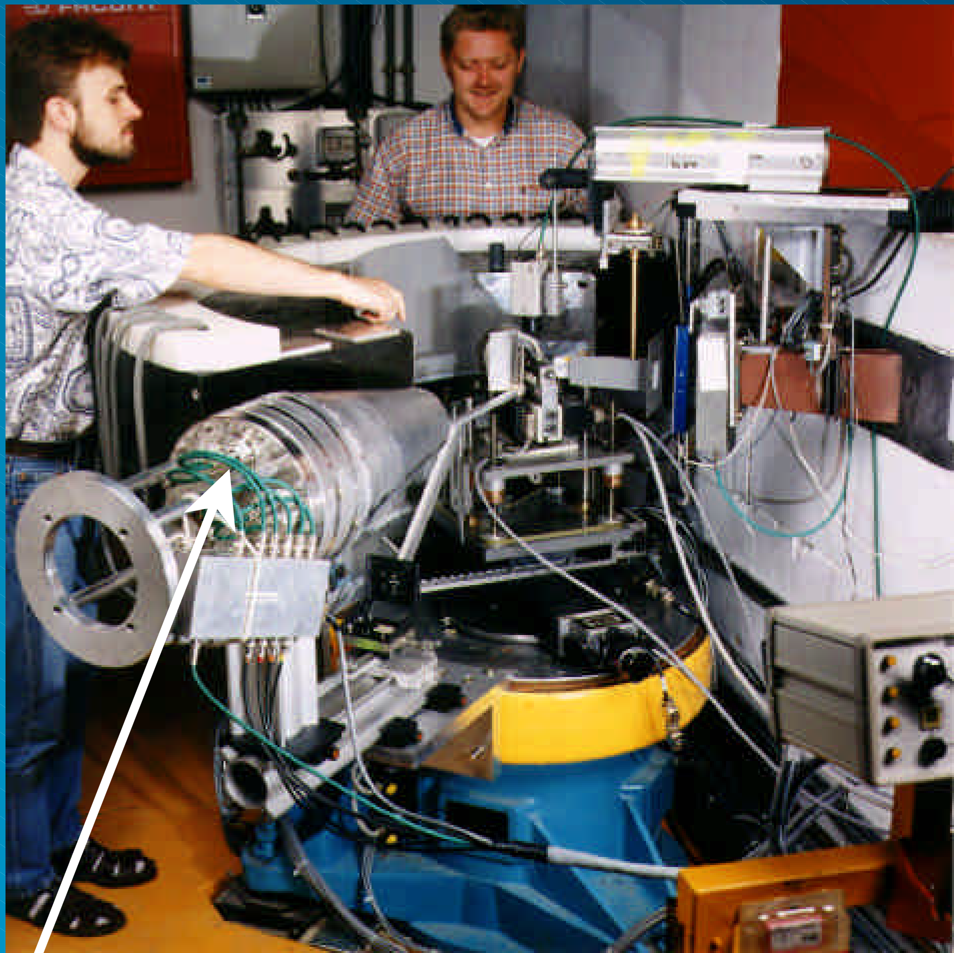


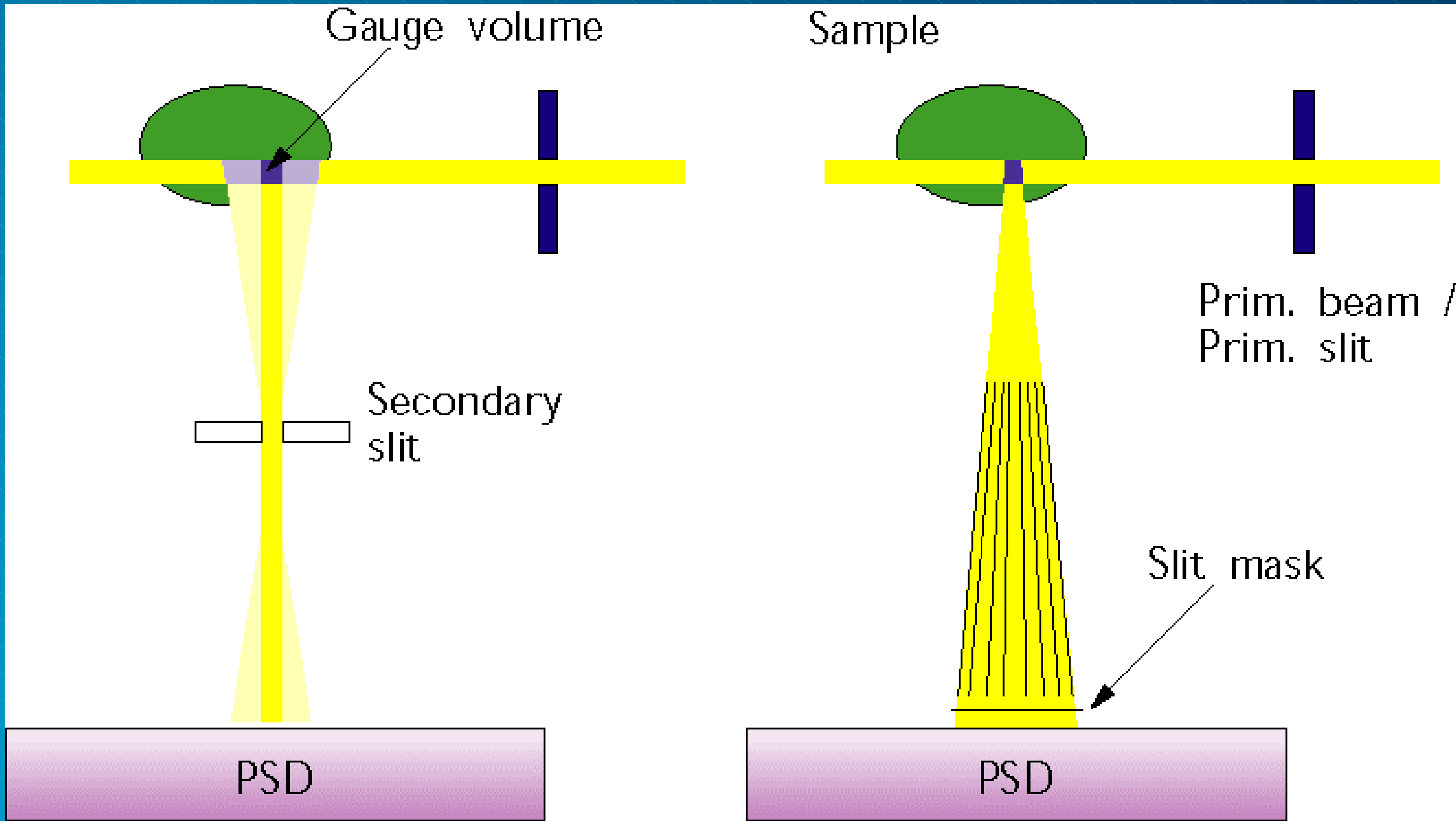
The 2D microstrip detector is used to obtain the complete line profile all at once

Neutron Strain Scanner

80x80 mm 2D Microstrip Detector on D1A

Thilo Pirling, Robert Wimpory

ILL Grenoble
Diffraction Group



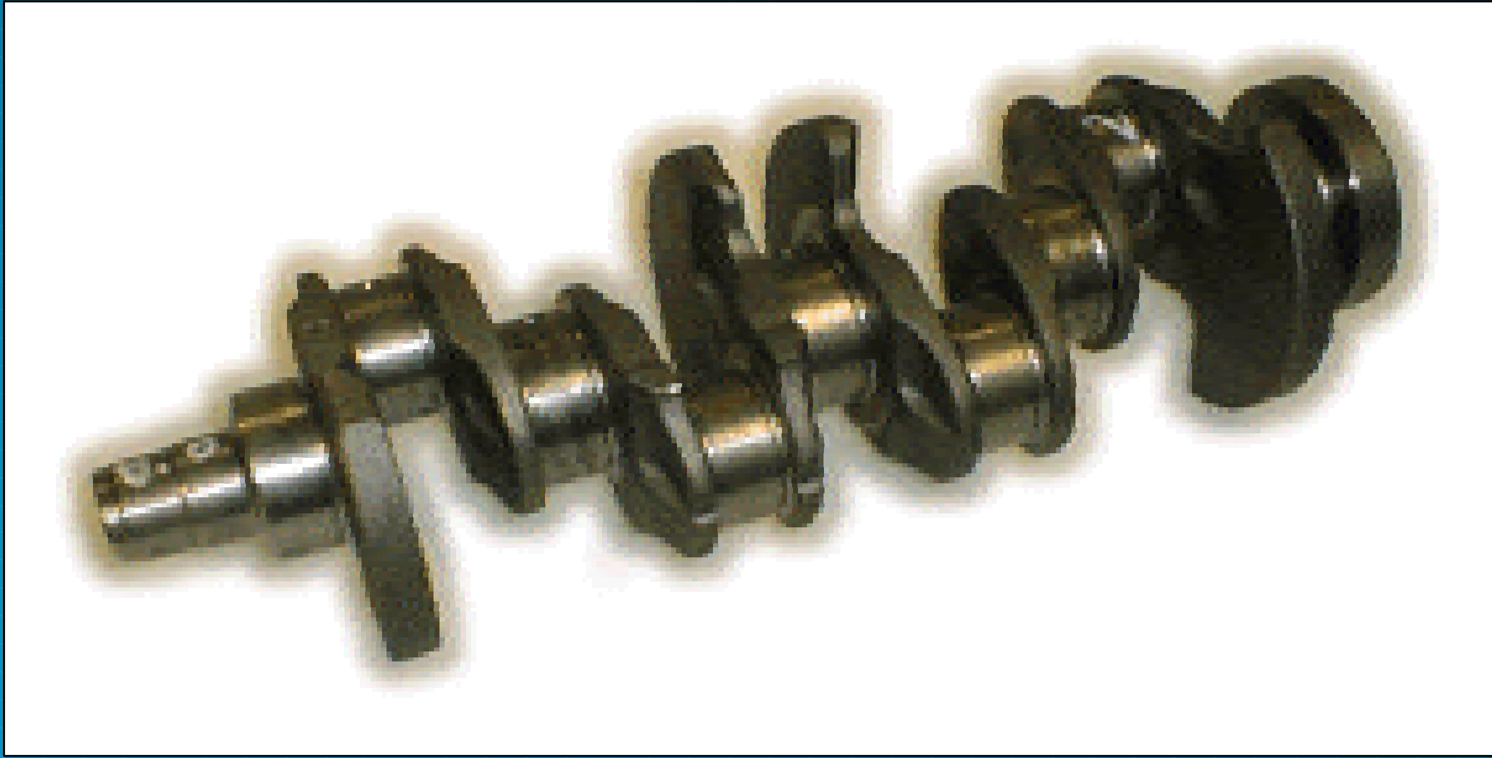
A large convergent collimator is used with the 2D microstrip detector
Note the very small sampling volume with this setup (right)

Neutron Strain Scanner

80x80 mm 2D Microstrip Detector on D1A

Thilo Pirling, Robert Wimpory

ILL Grenoble
Diffraction Group



The stress distribution in critical regions of this experimental crankshaft from Volkswagen was determined on the strain scanner at ILL.

ILL is part of the EU-RESTAND project with Volkswagen, Rolls-Royce, Airbus etc

A New ILL-EPSRC Strain Scanner

EPSRC grant of ~ 1M Pounds Sterling

Philip Withers (Manchester) et al., Thilo Pirling (ILL)

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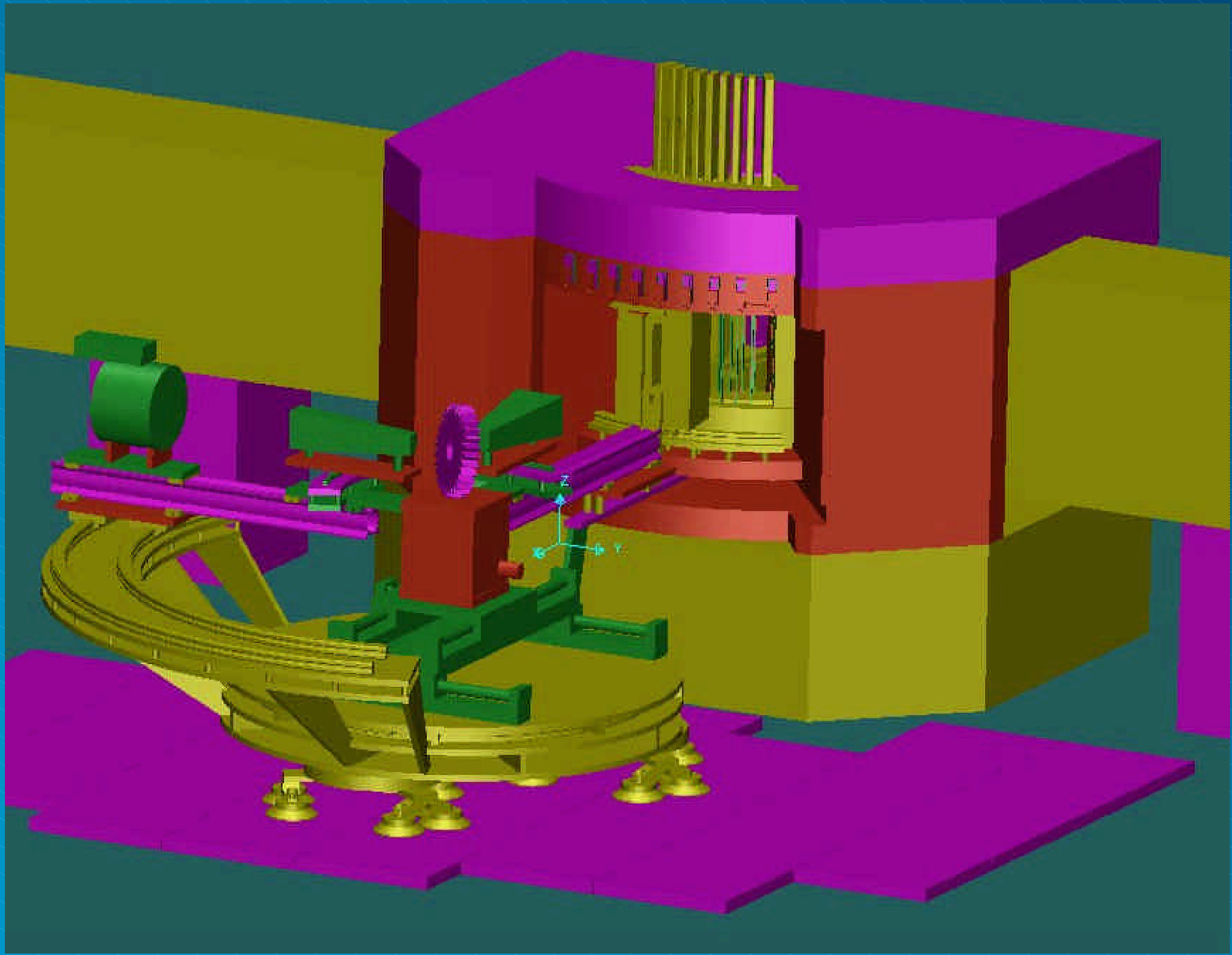

Artists impression of the new ILL-EPSRC strain scanner behind D1A/D1B

An Array of 2D Microstrip Detectors
D19 Fibre & Protein Diffractometer

Sax Mason, Trevor Forsyth, John Archer, Michael Walsh

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Diffraction Group

200x200 mm 2D microstrip detector for D19 fibre & protein diffractometer

An Array of 2D Microstrip Detectors

D19 Fibre & Protein Diffractometer

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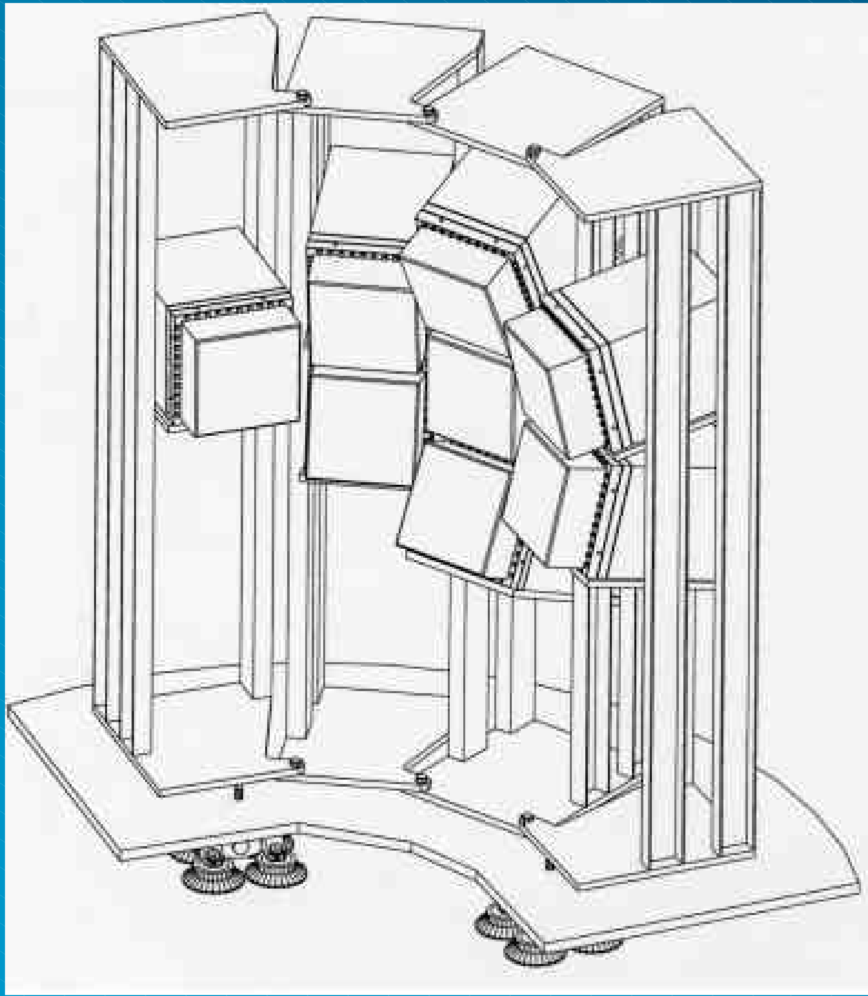
Sax Mason, Trevor Forsyth, John Archer, Michael Walsh



9 Independent 2D microstrip detectors

- 15 year old D19 detector covers only a thin 2D strip
- Replace with an array of high resolution 2D modules
- Increase efficiency x20
- Fibre Diffraction
  Small protein structures
  In-situ hydration studies.

Water in B-DNA sheets on D19

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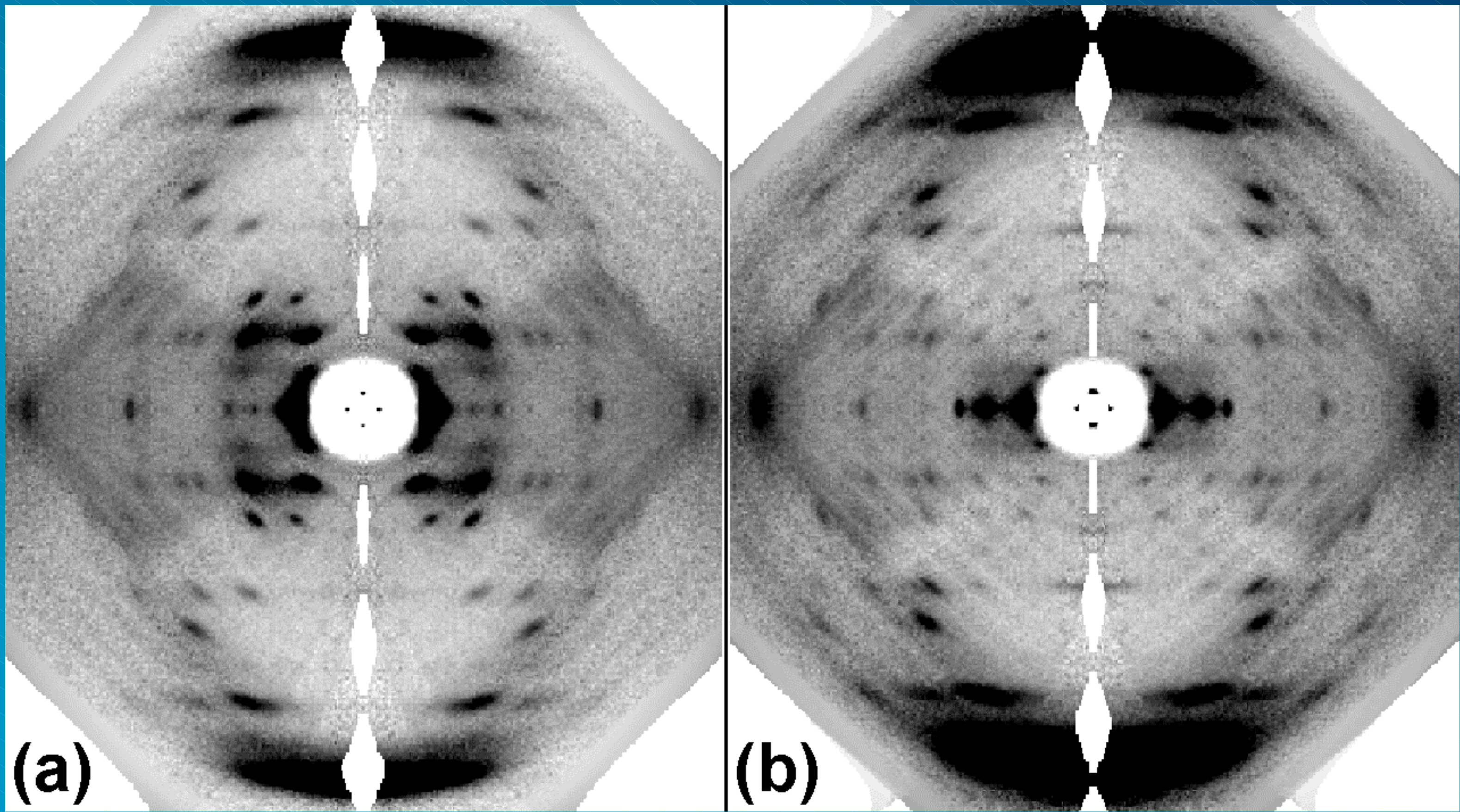
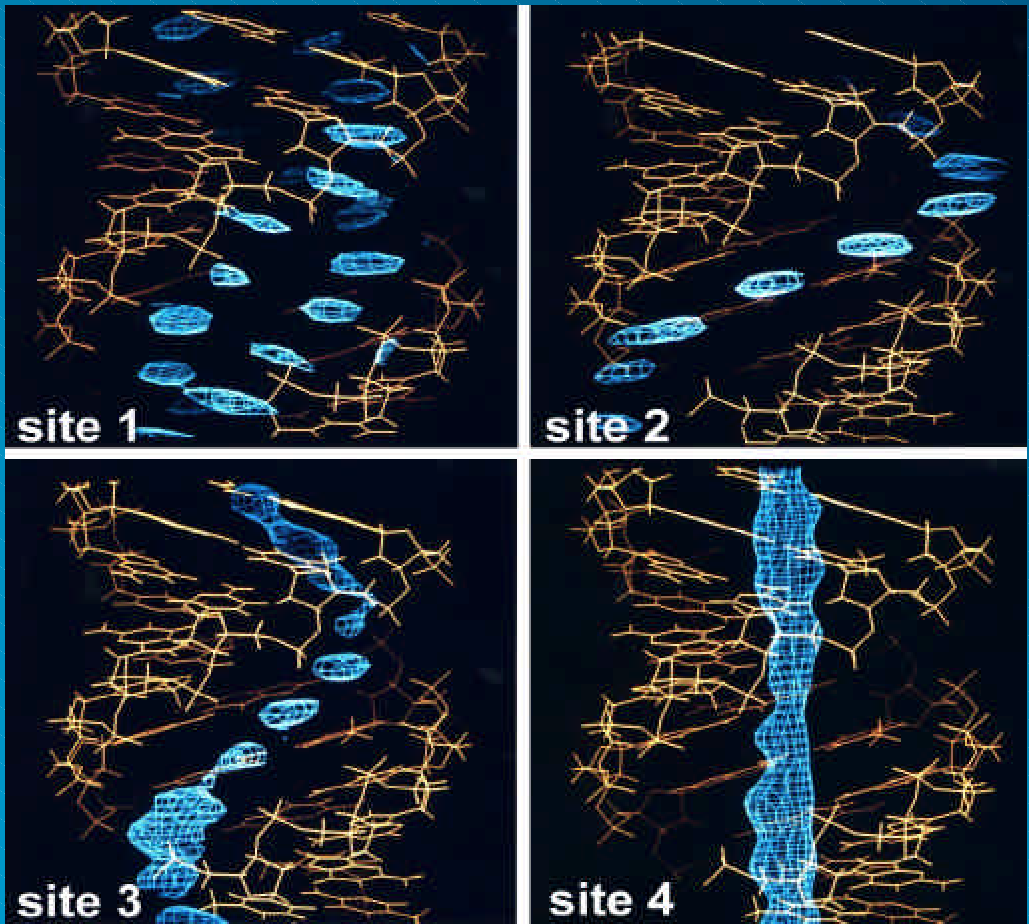
Shotton, Pope, Forsyth, Archer, Denny, Langan, Ye, Boote, (1998)
*J. Appl. Cryst.*31, 758



(a) with H_2O (b) with D_2O

Water in A-DNA Fibres on D19

Shotton et al, (1998) Biophys. Chem., 69, 8.
Pope et al, (1998) Physica B241, 1156.



- B-DNA sheets, but A-DNA fibres
- 100 individual DNA fibres in D_2O
- Diffuse fibre diffraction patterns from D19 used to locate water
- 4 distinct water sites located along double helix backbone

1) Bridging phosphate groups
2) Center of opening of major groove
3) Deep inside the major groove
4) Disordered string along helix axis

Why can't we do it with X-rays ?

Density of water in co-enzyme B12

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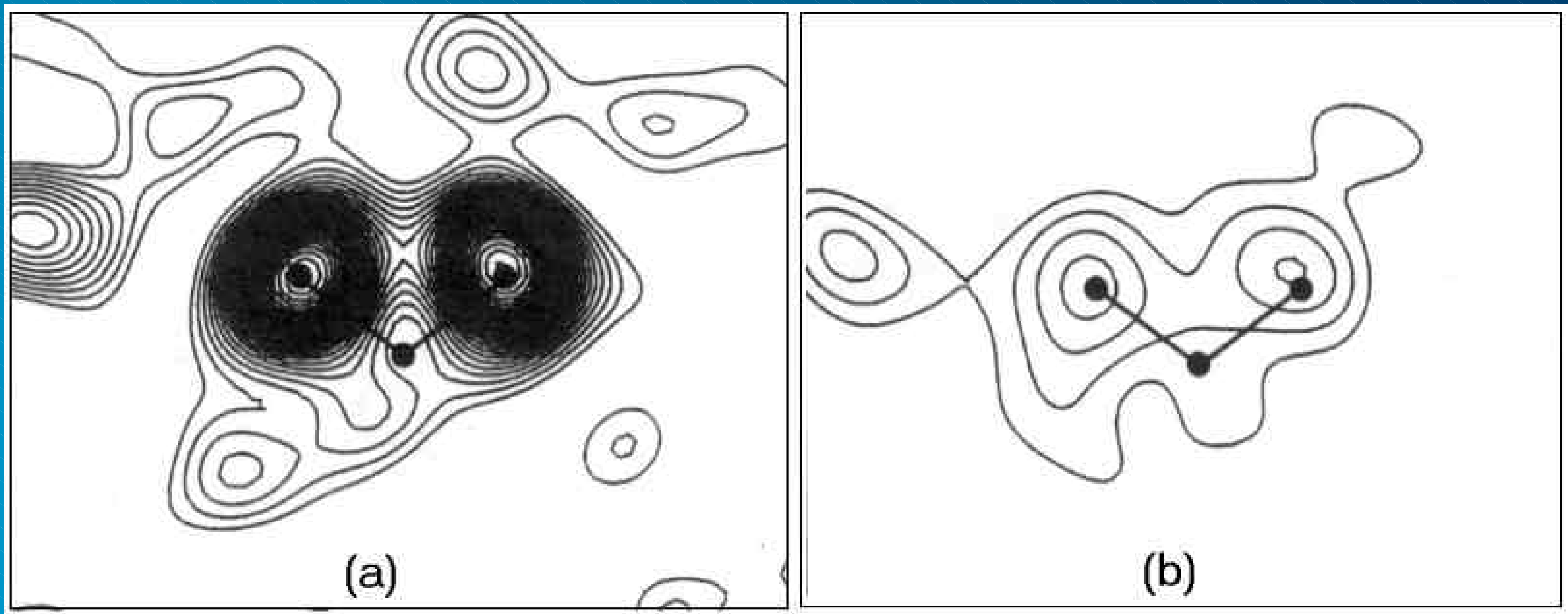
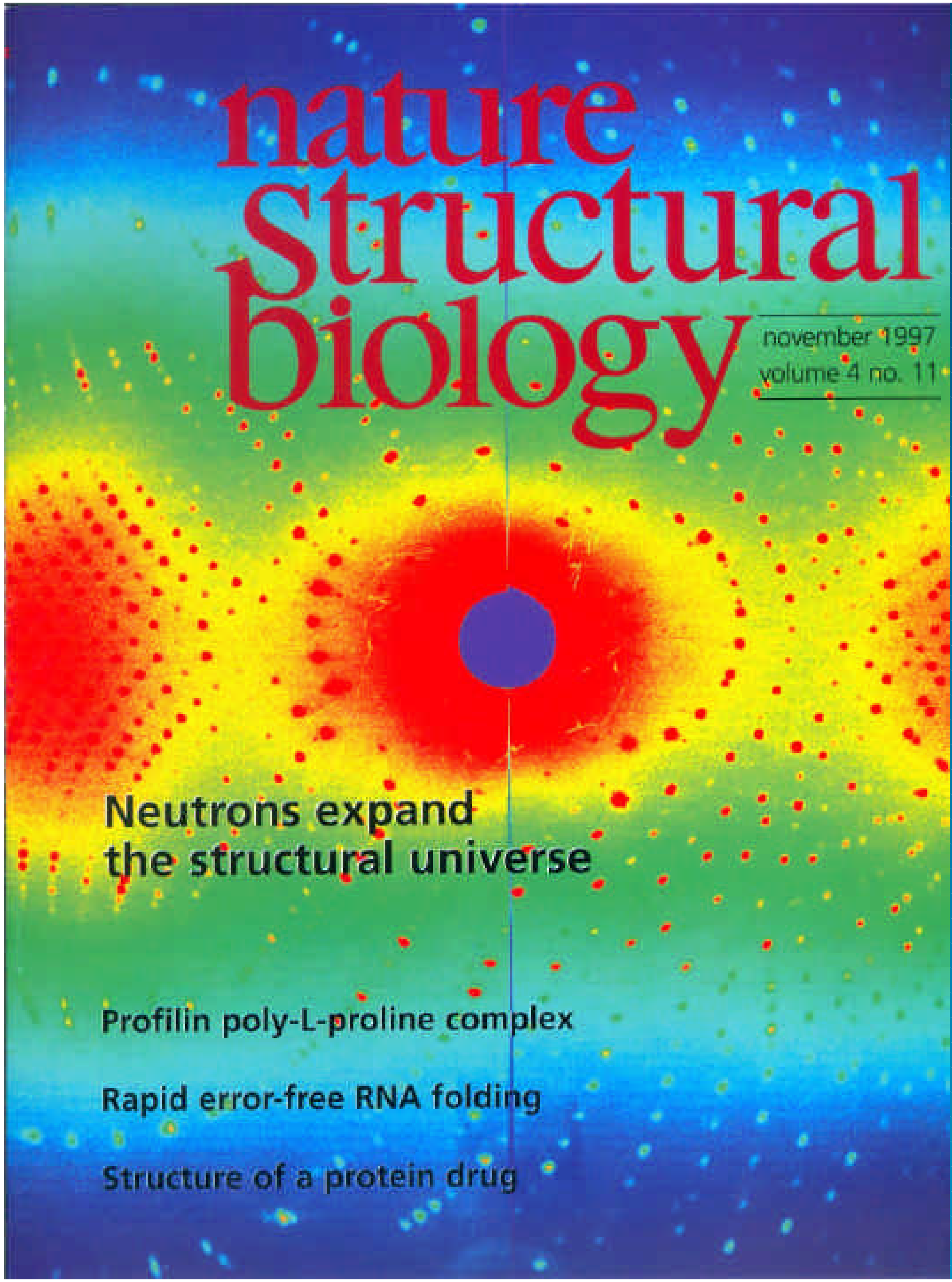
Langan, Lehmann, Wilkinson, Jogl, Kratky (1999) Acta Cryst D55, 51



D19 Neutron data

Synchrotron data



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Microstrip Detectors
vs
Neutron Image Plates

Nature (1997) Cover showing LADI data

(LAue Diffractometer with Image plates)

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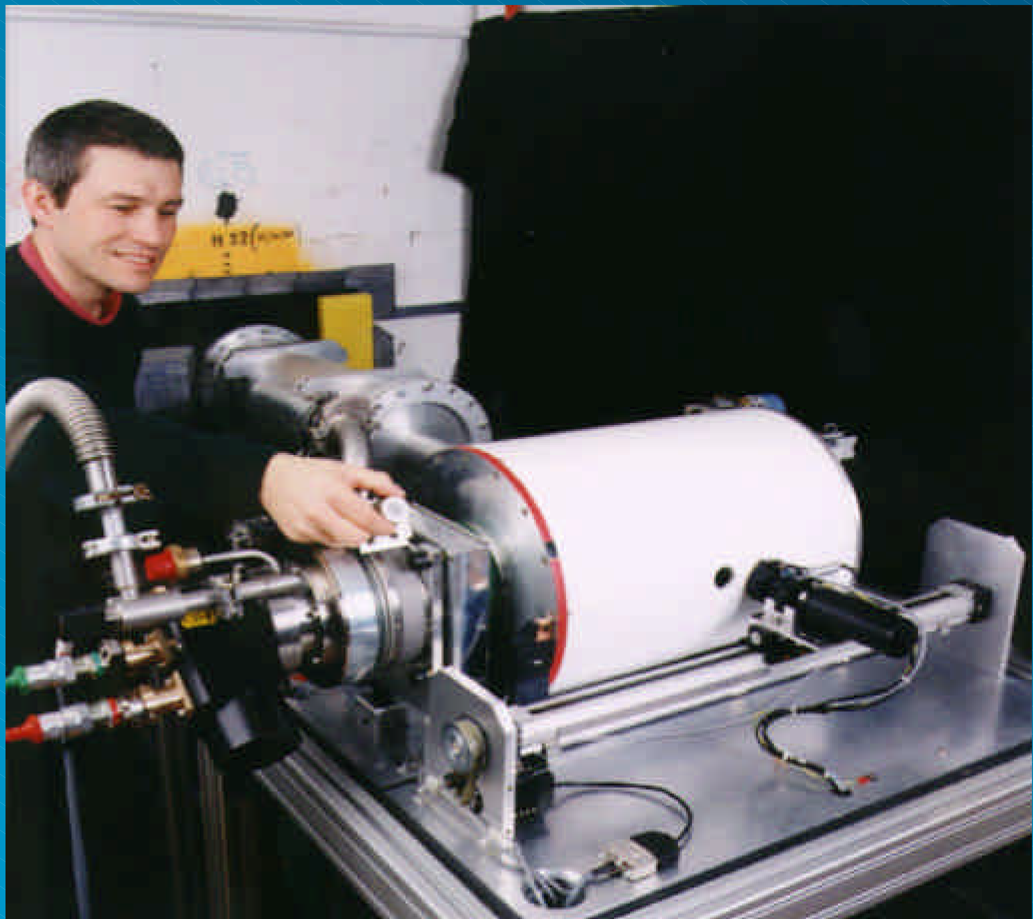
T-LADI Laue Neutron Image Plate for physics and chemistry

Dean Myles, Clive Wilkinson, Garry McIntyre



- Thermal neutron guide
- Band of neutron energies
- View reciprocal space
- In-situ laser readout
- Unique survey of P/T
- Phase T/Ns, superstruct.

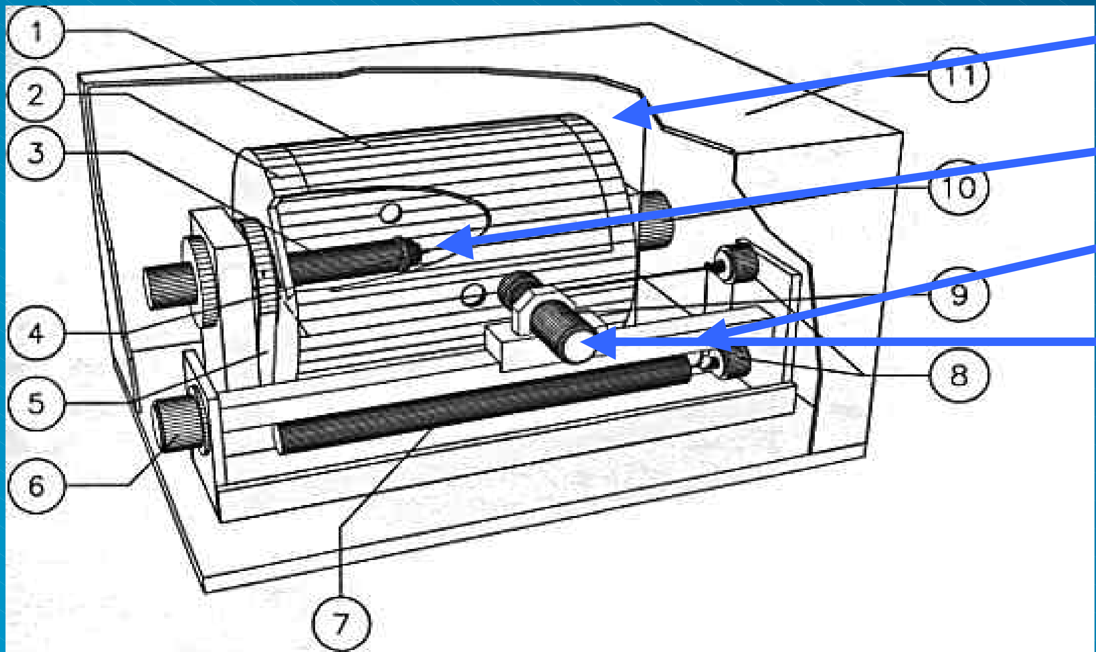
Dean Myles with LADI and cryo-refrigerator on thermal guide H22

T-LADI Neutron Image Plate for physics and chemistry

Dean Myles, Clive Wilkinson, Garry McIntyre

ILL Grenoble

Diffraction Group



1. Image plate on rotating drum

3. Sample holder

7. He-Ne laser

9. Reader head, photomultiplier

Phonograph readout time 4 min.

4000x2000 pixels of 200 μm

Original LADI (used for biological structures) adapted for materials research

T-LADI Neutron Image Plate
Superstructure in $La_2Co_{1.7}$

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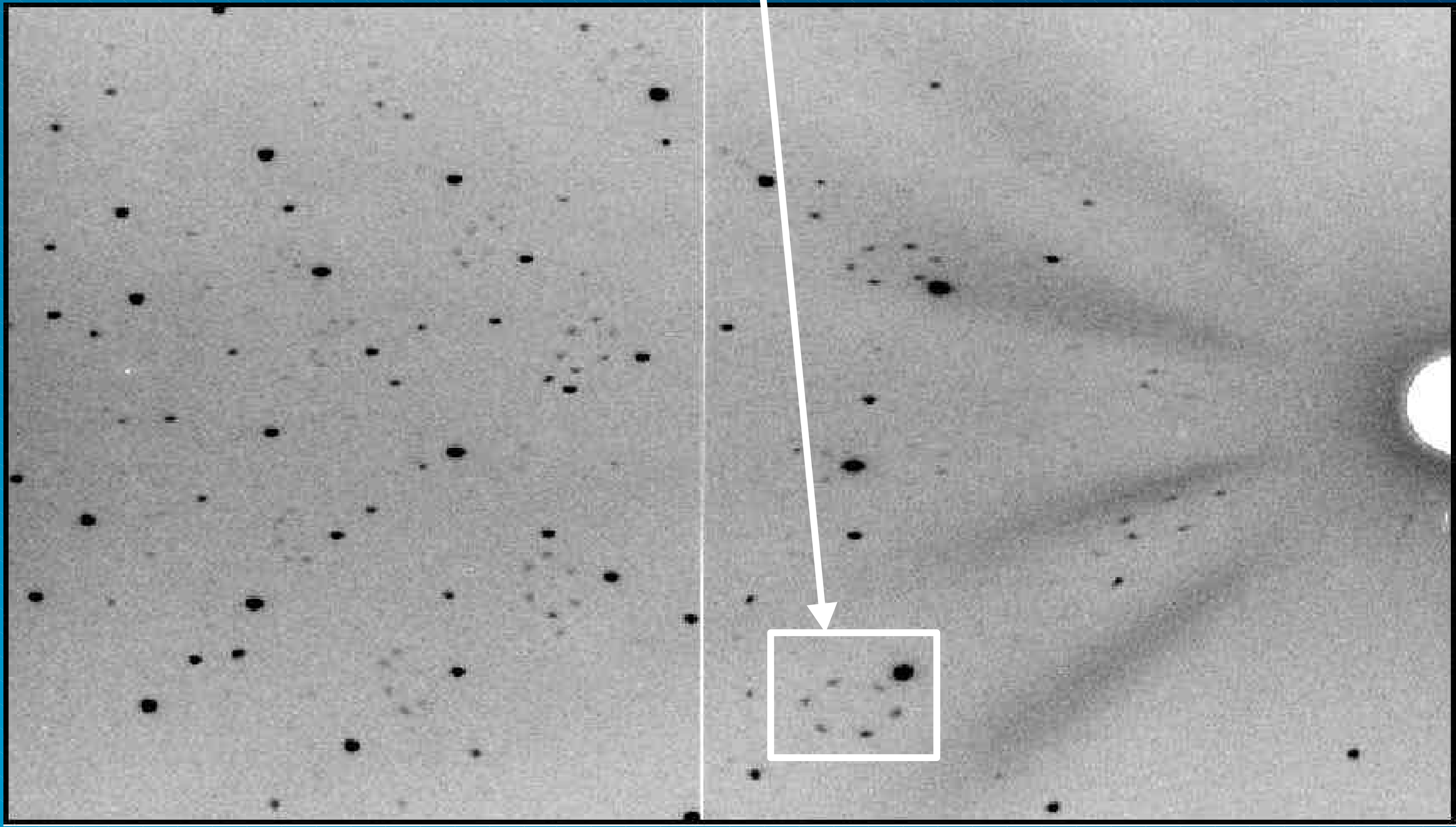

$La_2Co_{1.7}$ on T-LADI showing incommensurable superstructure

T-LADI Neutron Image Plate
5-fold symmetry of quasi-crystal

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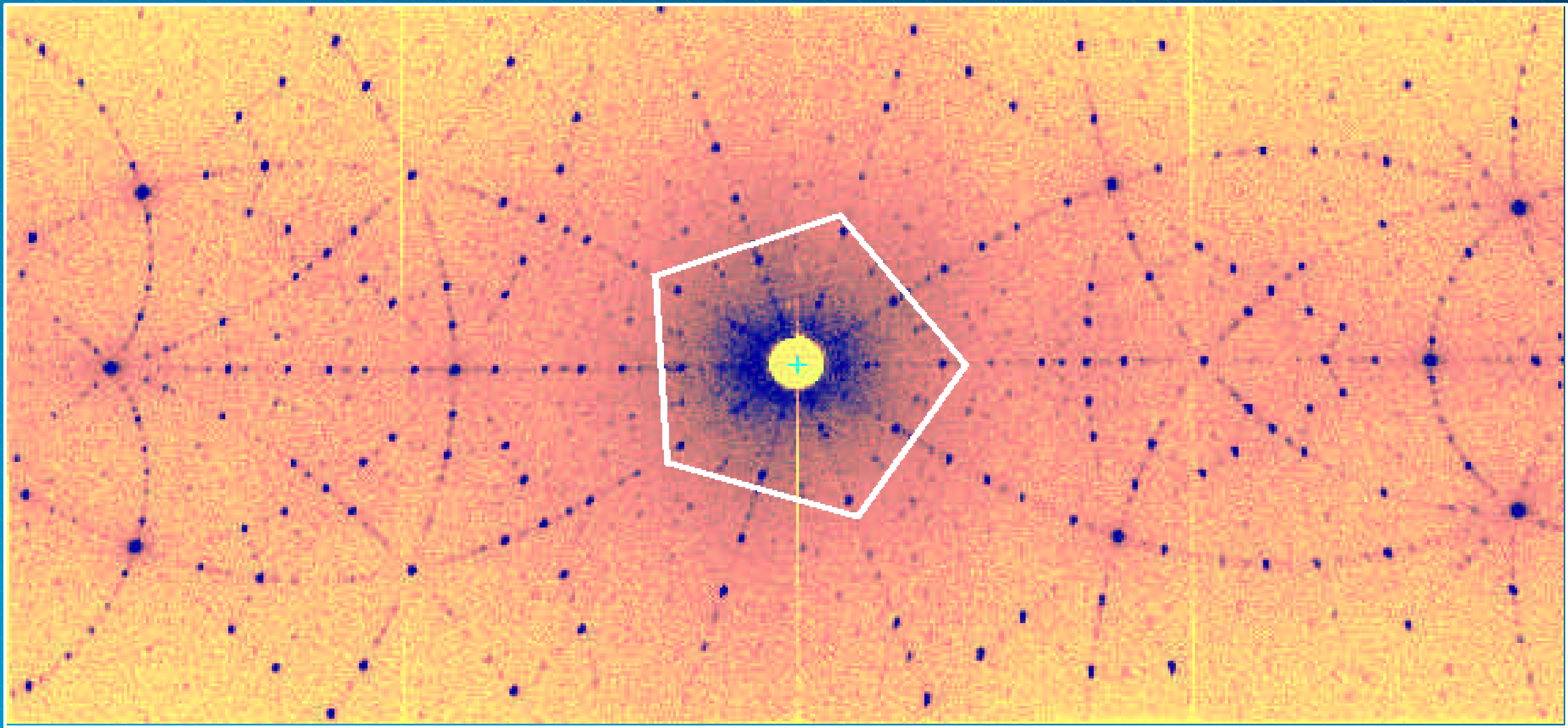
5-fold symmetry axis in ZnMgY quasi-crystal - De Boissieu et al. (1999)



T-LADI neutron image plate photo courtesy of G. McIntyre, Oct 1999

T-LADI Neutron Image Plate
5-fold symmetry of quasi-crystal

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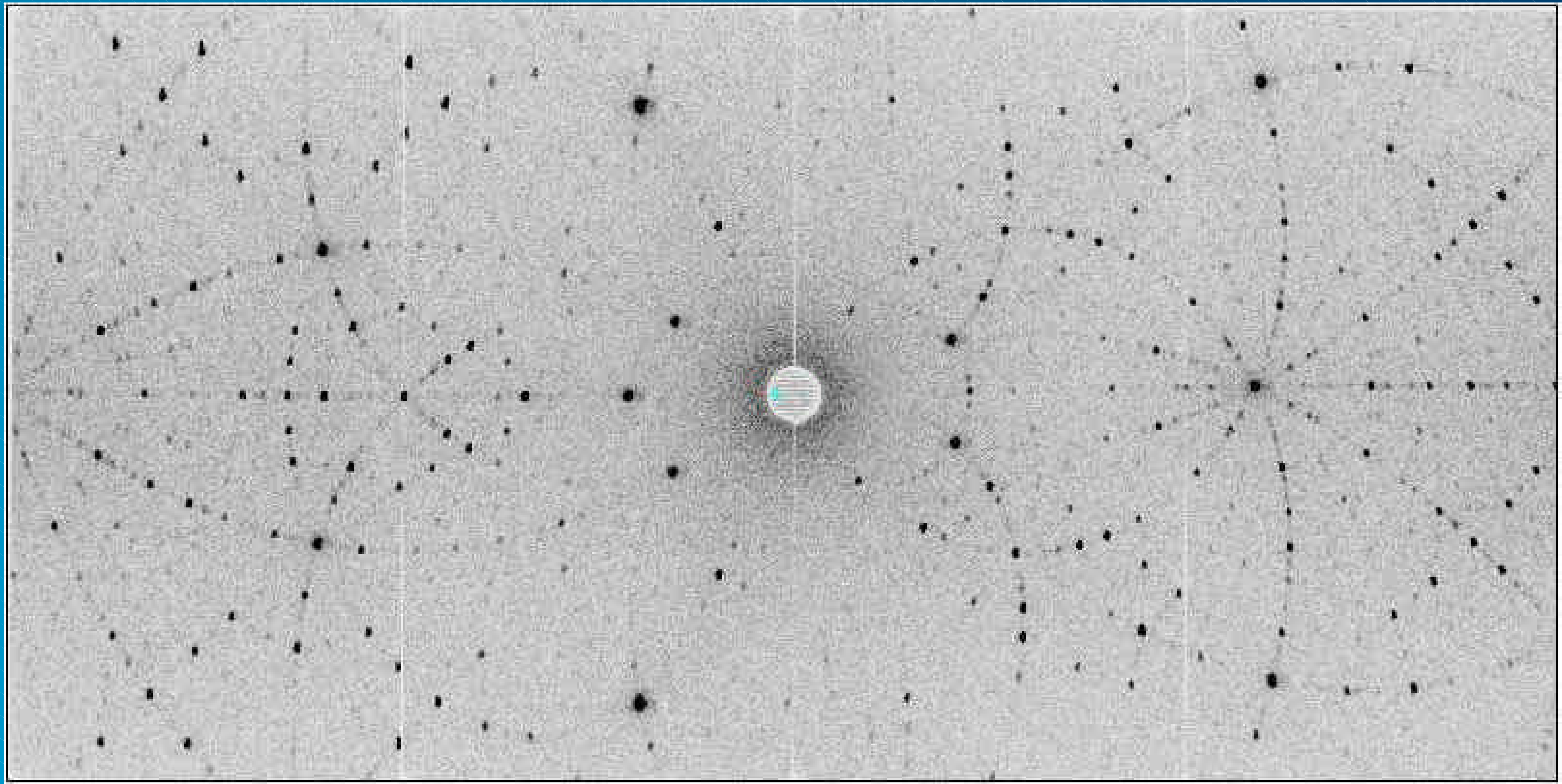
Rocking the ZnMgY quasi-crystal (Dynamics) – McIntyre, Cowan (1999)



T-LADI Neutron Image Plate
Why Image-plates + Microstrips ?

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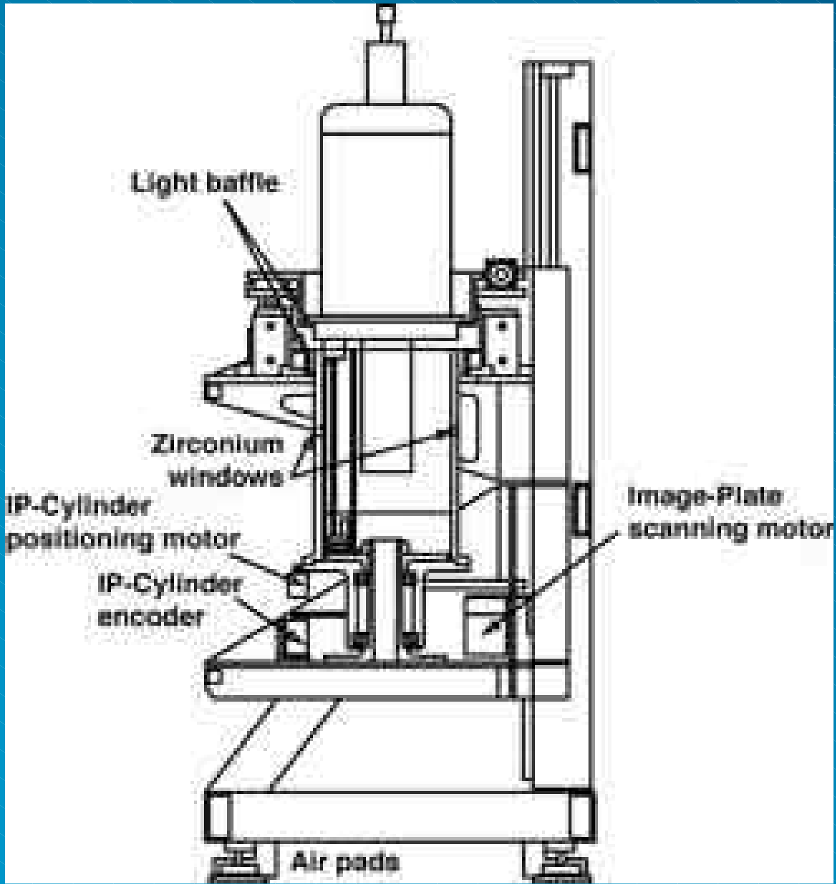

Disadvantages of Image-plates

- Photographic technique
- Accumulate background
- Background from all λ (wide Δλ)
- H-background

For X-rays, photographic techniques are now replaced by electronic PSD's

New T-LADI uses thermal neutrons, more efficient interior read-out optics, vertical geometry allowing use of cryostats, furnaces, magnets, pressure cells

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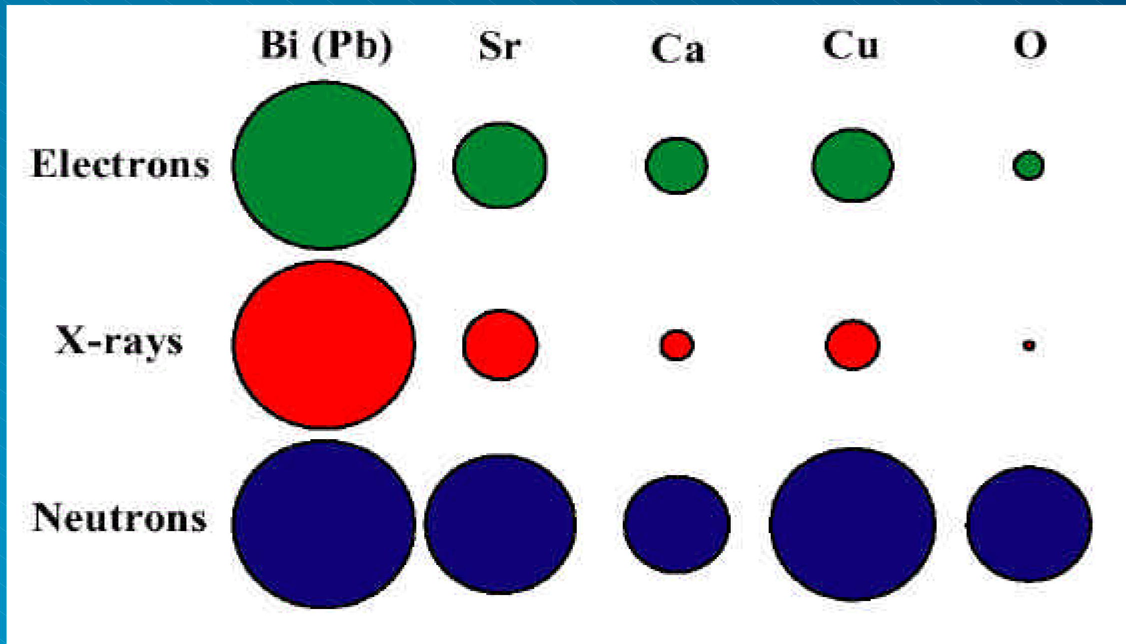
Why Neutrons ?

- Strong Magnetic Scattering of Neutrons
- Relative Scattering Powers of the Elements



- Neutrons scatter strongly from light elements
- Neutrons scatter strongly at high angles (resolve)

Impact of neutron powder diffraction

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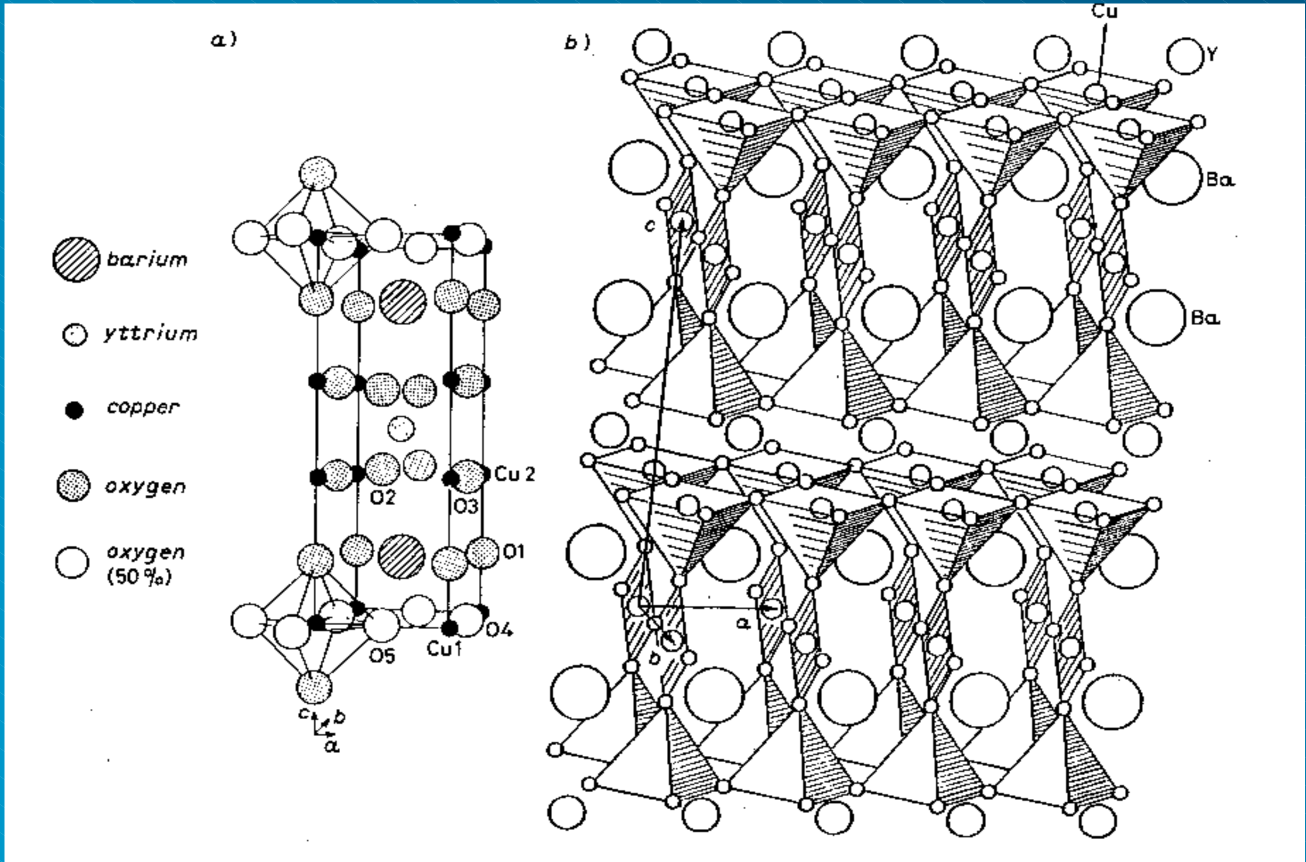
Heavy metal oxides are still with us - Superconductors, GMR



- Structure of the 90K high Tc superconductor
  - Left - by X-rays (Bell labs & others)
  - Right - by Neutrons (many neutron labs)
- The neutron picture gave a very different idea of the structure - important in the search for similar materials.

$YBa_2Cu_3O_7$ drawing from Capponi et al 1987 (2nd most cited ILL paper)

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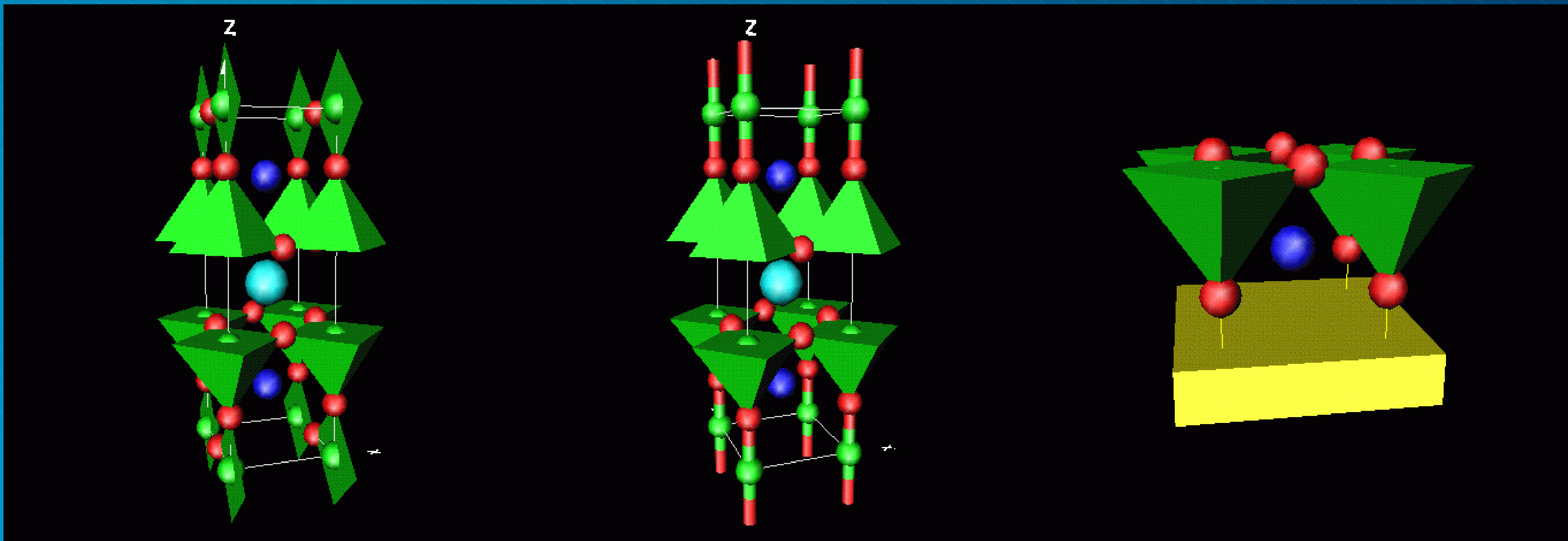
Neutron Powder Diffraction
Essential technique for new materials

Most cited ILL paper - “charge reservoir” concept in oxide superconductors

- **Superc. $YBa_2Cu_3O_7$**
- **Non-superc. $YBa_2Cu_3O_6$**
- **Charge Reservoir**



- Cava, R. J. et al. (1990). Physica C. **165:** 419 (Bell labs/CNRS/ILL)
- Jorgensen, J.D. et al. (1990) Phys.Rev. **B41**,1863 (Argonne)

Electronic Order-Disorder

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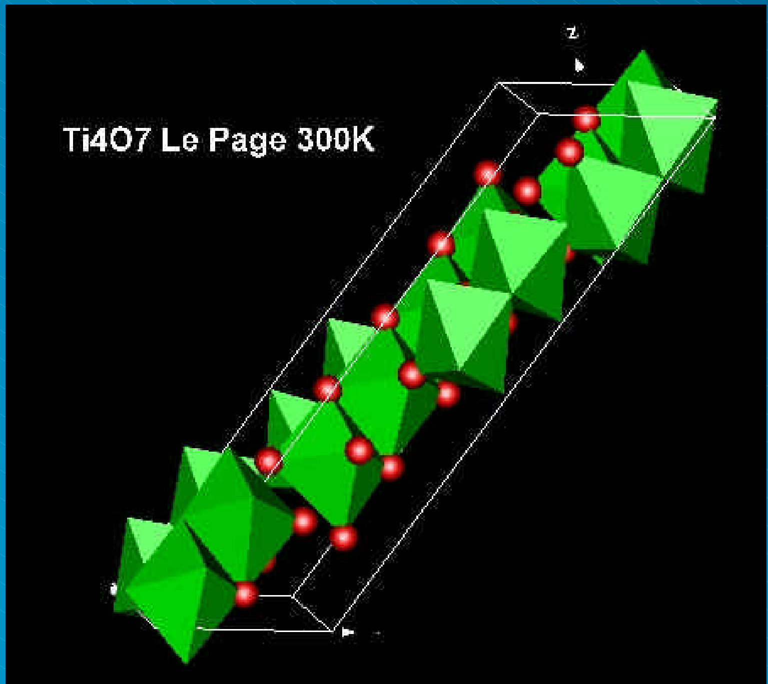
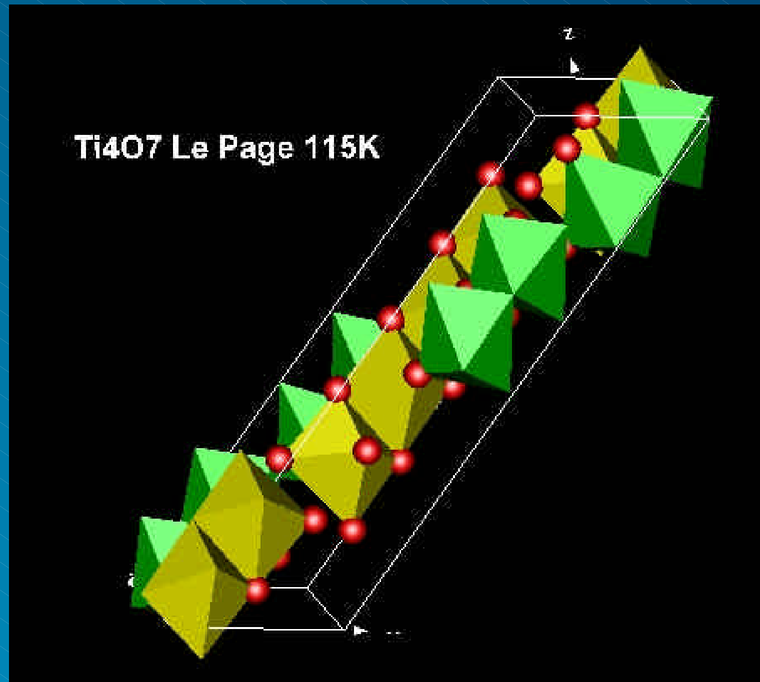
- Oxide superconductors, CMR, Vewey transition…
- Precise structural measurements vs temperature





- Example: charge ordering in Ti_4O_7 (Le Page et al.

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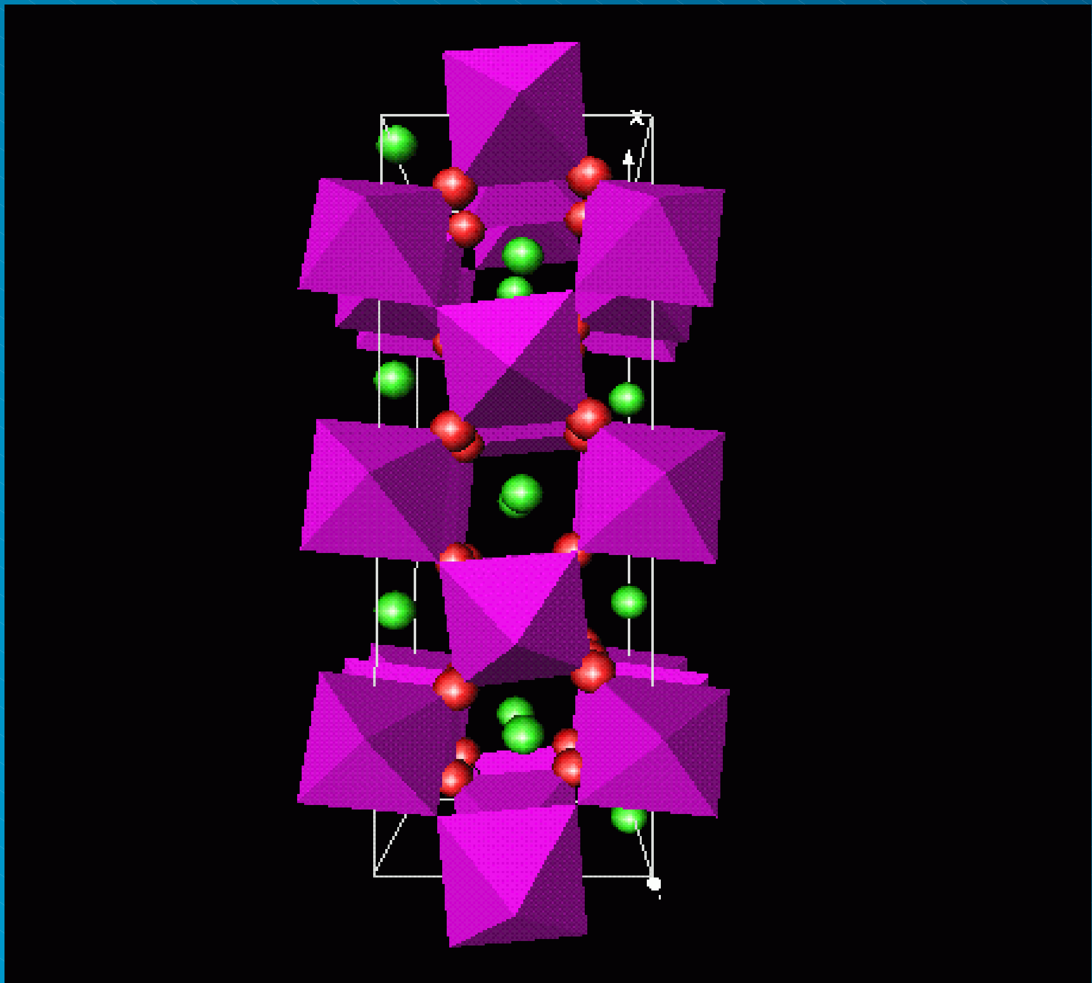
Giant Magneto-Resistive Ceramics
$La_{0.333}Ca_{0.667}MnO_3$ on D2B



- Very large changes in electrical resistivity with temperature
- cf oxide superconductors
- Mixed valence charge-ordering Mn^{3+}/Mn^{4+}
- GMR effect near room temperature
- Applications to magnetic storage of data (new high density IBM hard disks)

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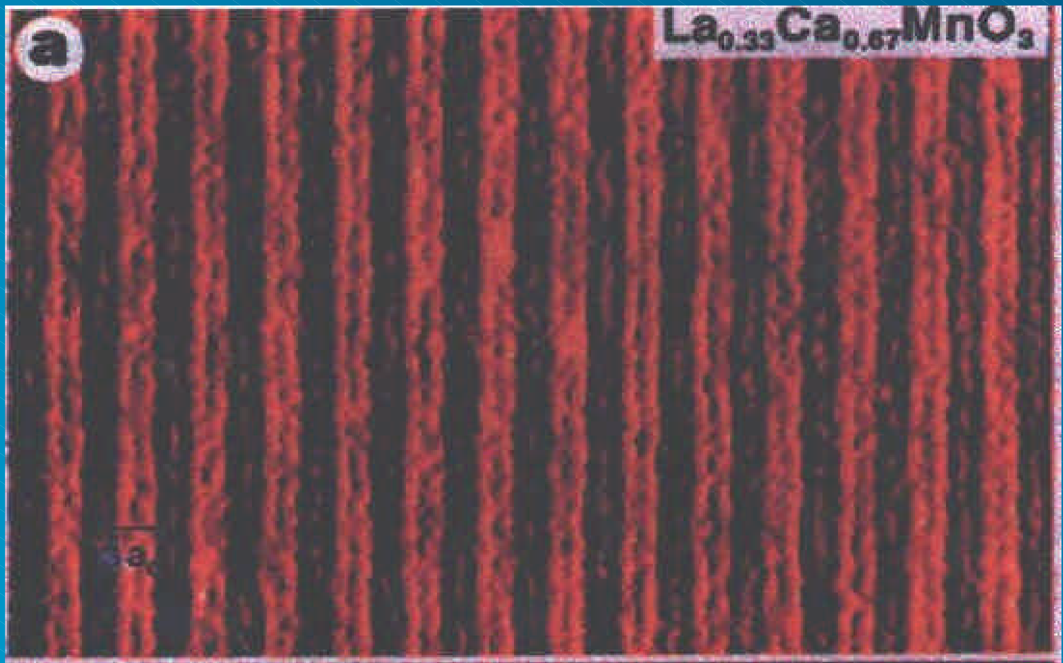
GMR Stripes and Charge Ordering

1D-ordering ? Dimensionality important for theory.



Mori et al. Nature (1998) 392,473
Other papers in Phys. Rev. Letters

- **Remarkable electron microscope images of 1D stripe pattern in GMR $La_{0.33}Ca_{30.67}MnO_3$**
- **Evidence also for 1D ordering in high-Tc superconductors (Cu^{3+} stripes, spin-ladders etc)**

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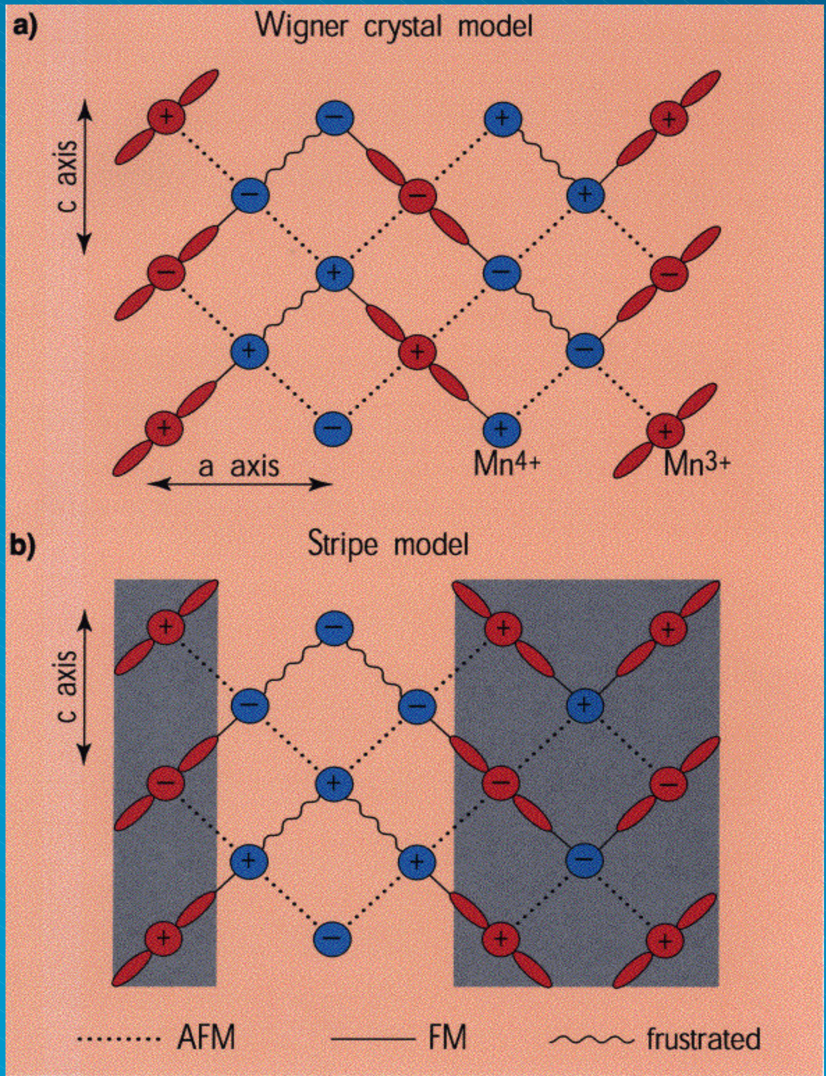
GMR Stripes and Charge Ordering

1D-ordering ? Dimensionality important for theory.



- **Expect instead Mn^{3+}/Mn^{4+} to be uniformly distributed (2D Wigner crystal model of Goodenough)**
- **The 1D-stripe model would have very important consequences for the theory of superconductors and GMR oxides**

GMR Stripes and Charge Ordering

Neutron + Synchrotron Powder Diffraction

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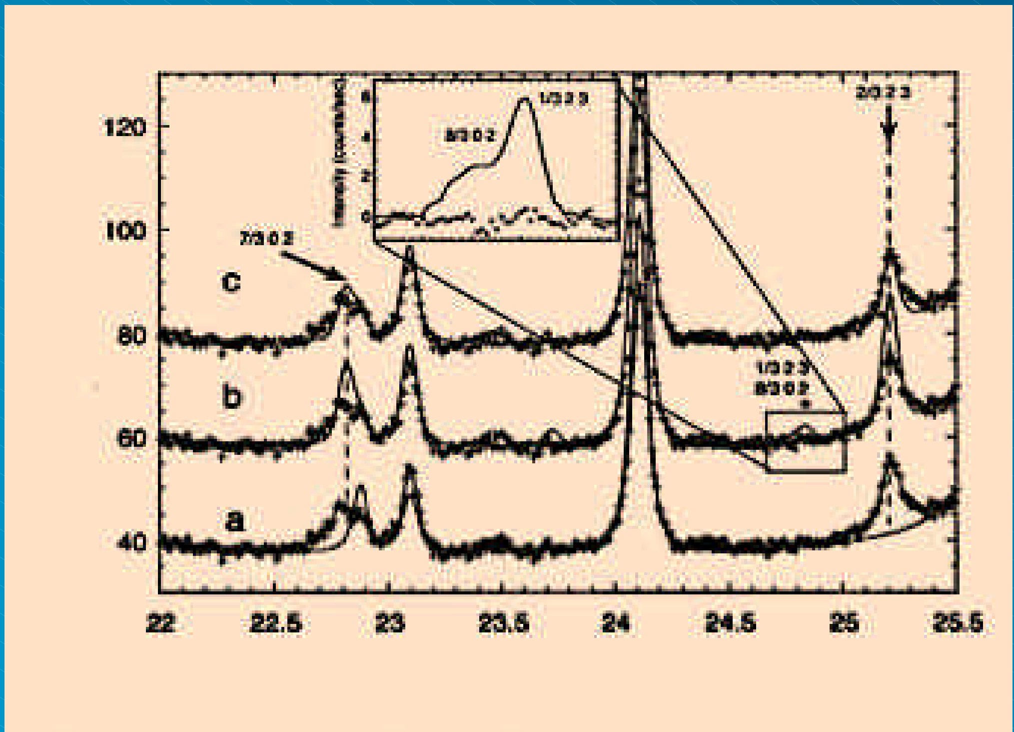

Radaelli et al. (1999) Phys. Rev B
X-ray work on X7A (BNL)
Neutron work on D2B (ILL)

- **High resolution synchrotron powder data (Brookhaven) reveals true symmetry.**
- **High resolution neutron powder data (ILL Grenoble) allows refinement of real structure.**

  a) Average Structure
  b) Stripe Structure
  c) Wigner Crystal Structure (best fit)

- **<u>The stripe structure is not supported</u>**

Early Days at ILL Grenoble (1972)

First ILL Powder Diffractometer D1a

- Small soller collimator
- Single detector
- Shared monochromator
- -High Resolution, BUT
  -Very Low Intensity

Second Generation Machines (1984)

High Resolution with Very Large Detector bank (D2B)

- 64 High Resolution Plastic Foil Collimators
- Large Composite Focussing Monochromator
- High Resolution
- Good Intensity

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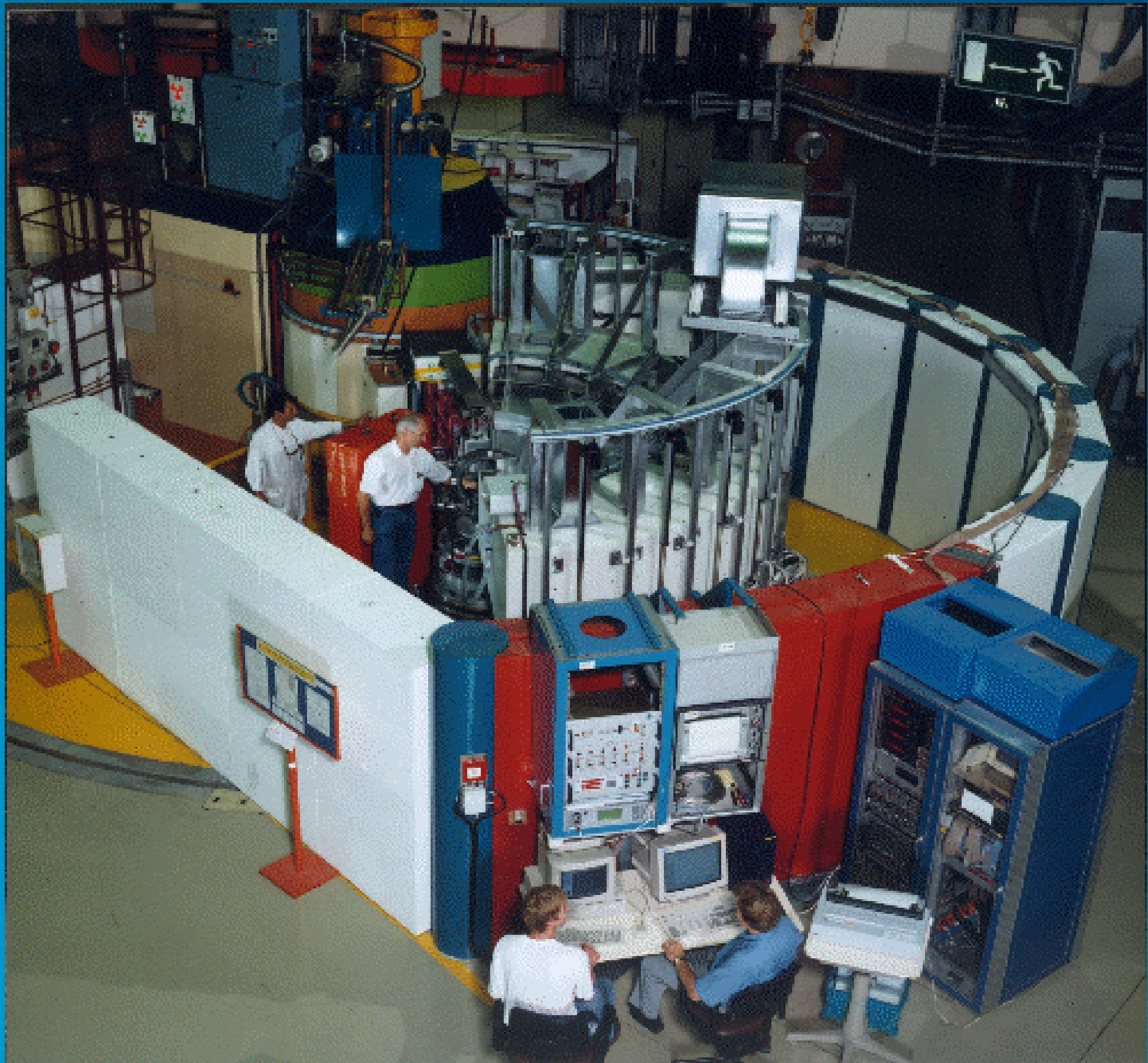
High Flux Powder Diffractometer D20

Diffraction Group

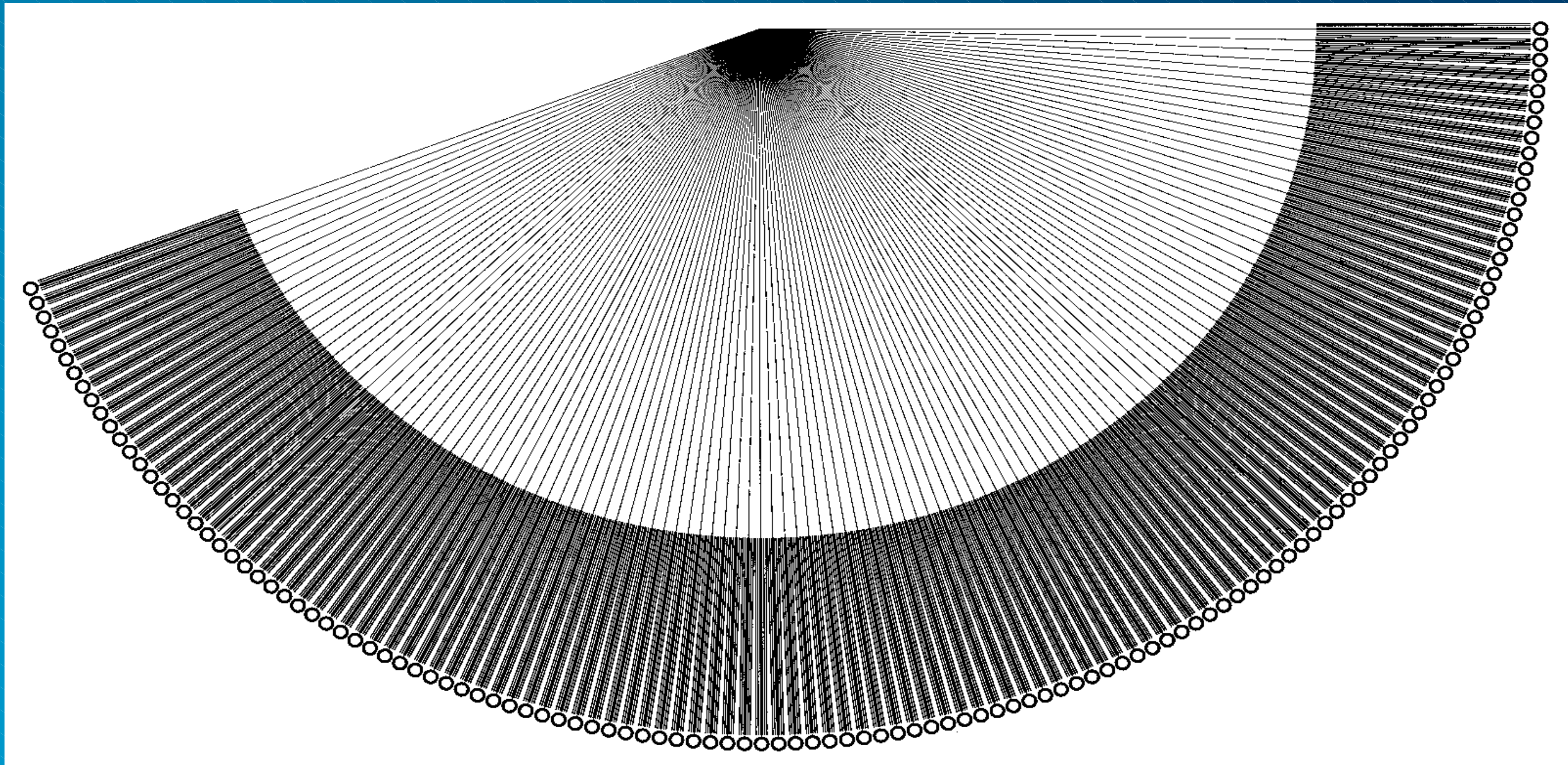
Pierre Convert, Thomas Hansen, Jacques Torregrossa



D20 in action with
Jacques Torregrossa,
Pierre Convert
& Thomas Hansen

The Future - Big Detectors

Large pseudo-2D PSD (array of linear-wire detectors)



- 2D detector allows both high efficiency & high resolution

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The Future - Big Detectors

- 128 linear wire PSD detectors, Y-resolution 1^{o}, height 300 mm
  Cost **1.25 MFF** (available commercially from 2 sources)

- 128 high resolution soller collimators, X-resolution 5’, 300 mm
  Cost **1.25 MFF** (International tenders 1999, prototype produced)

- New detector protection, B_4C-epoxy
  Cost **~0.5 MFF** (local company)

- **Total 3 MFF (0.75 M$A)**

Polarized Neutrons & He^3 Filters

Francis Tasset, Eddy Lelievre, Adrin Hiller, Trefor Roberts

ILL Grenoble

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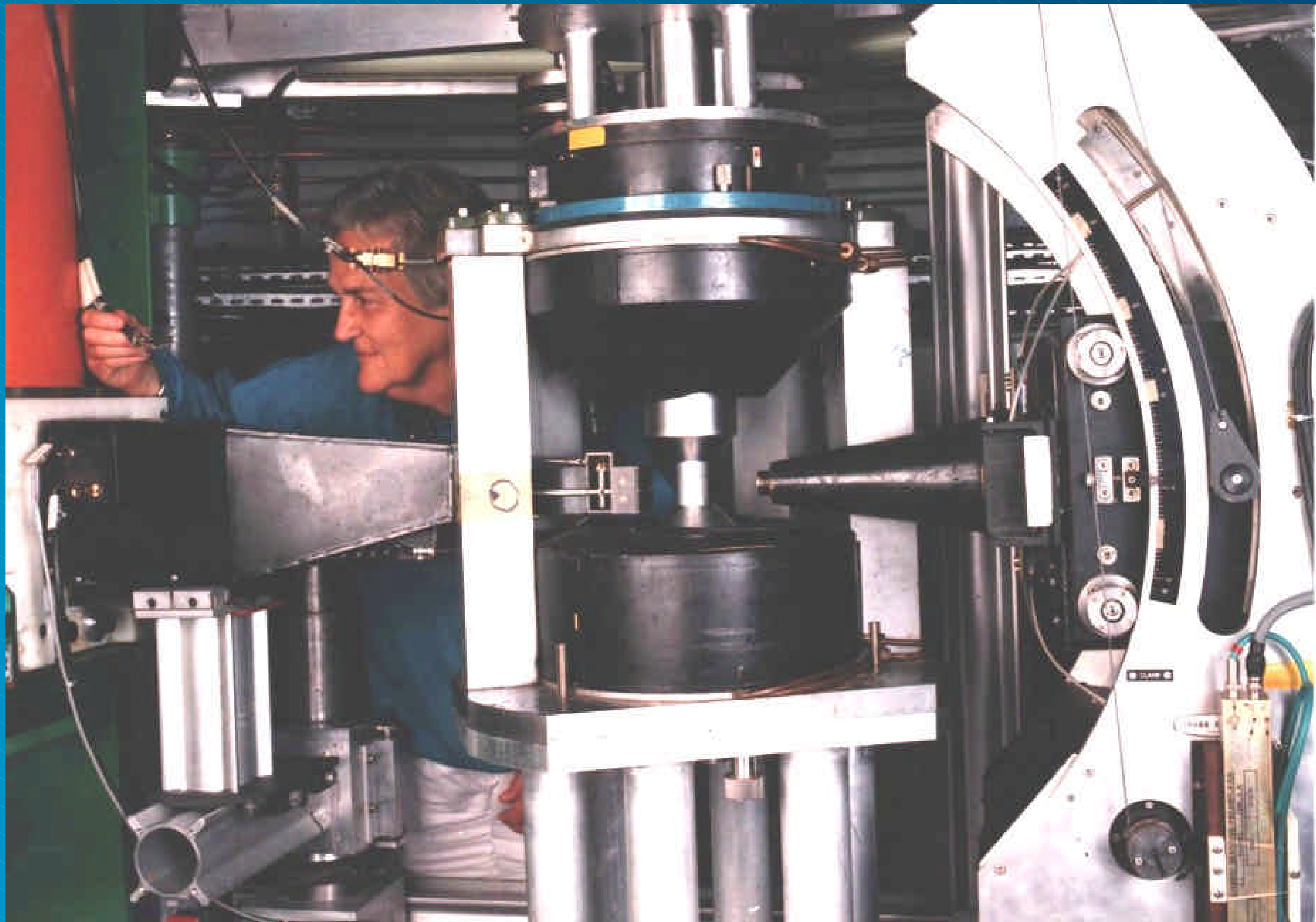
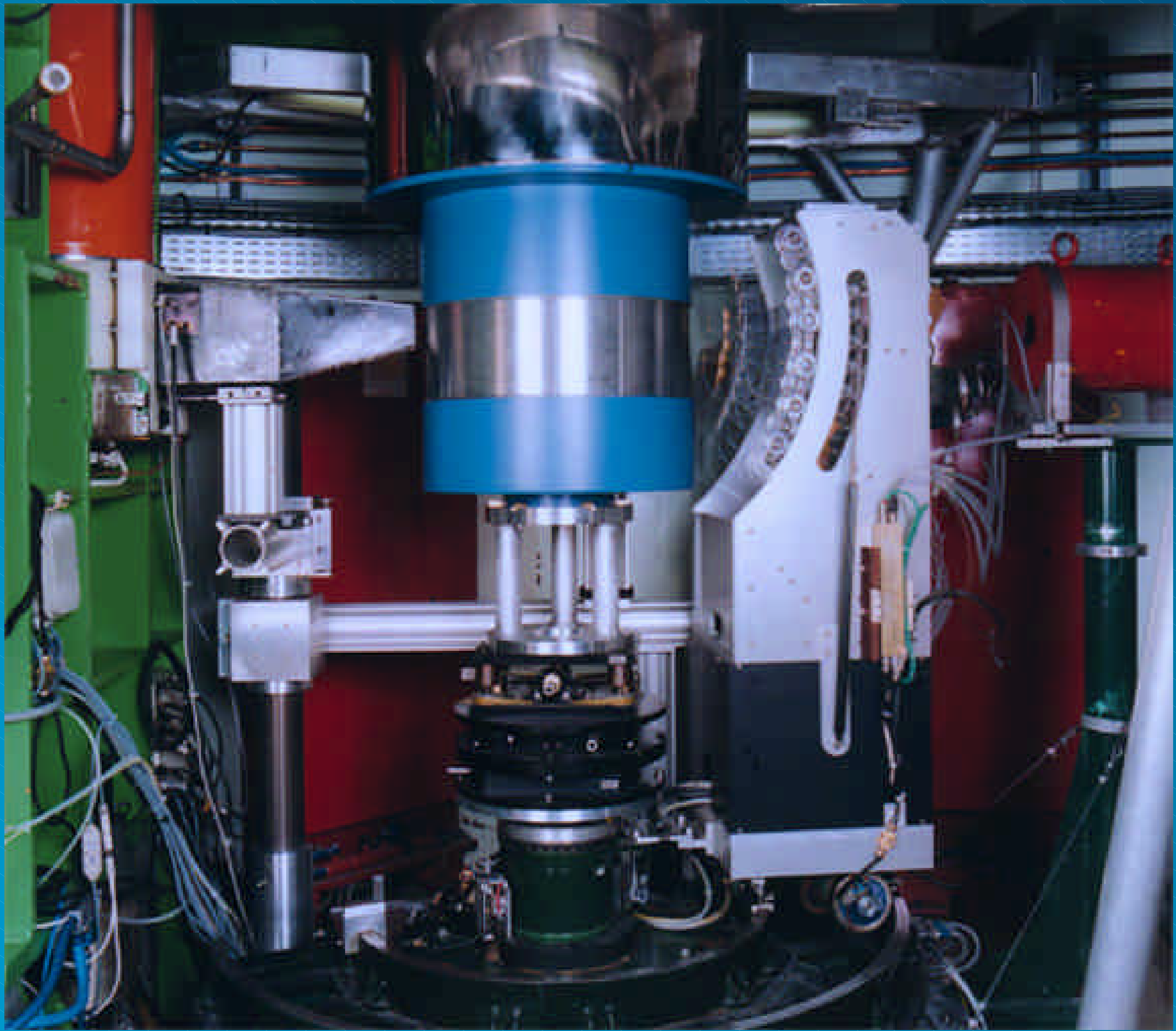


Jane Brown with magnet on D3

Polarized Neutrons & He^3 Filters

Francis Tasset, Eddy Lelievre, Adrin Hiller, Trefor Roberts

ILL Grenoble
Diffraction Group



New 10 Tesla cryo-magnet with lifting counter on D3

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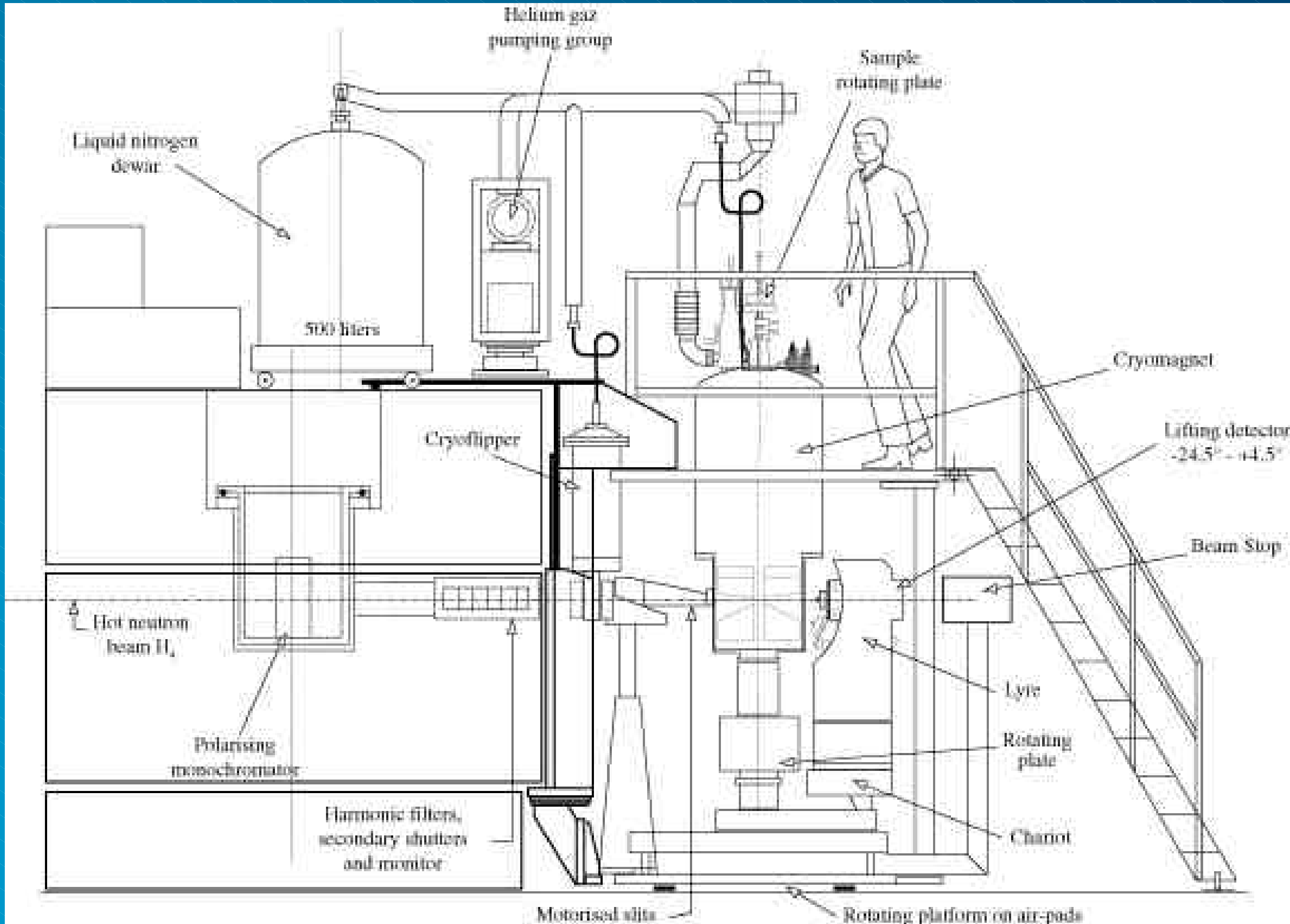
D3 Polarized Neutron Diffractometer

Francis Tasset, Eddy Lelievre, Adrin Hiller, Trefor Roberts

Diffraction Group



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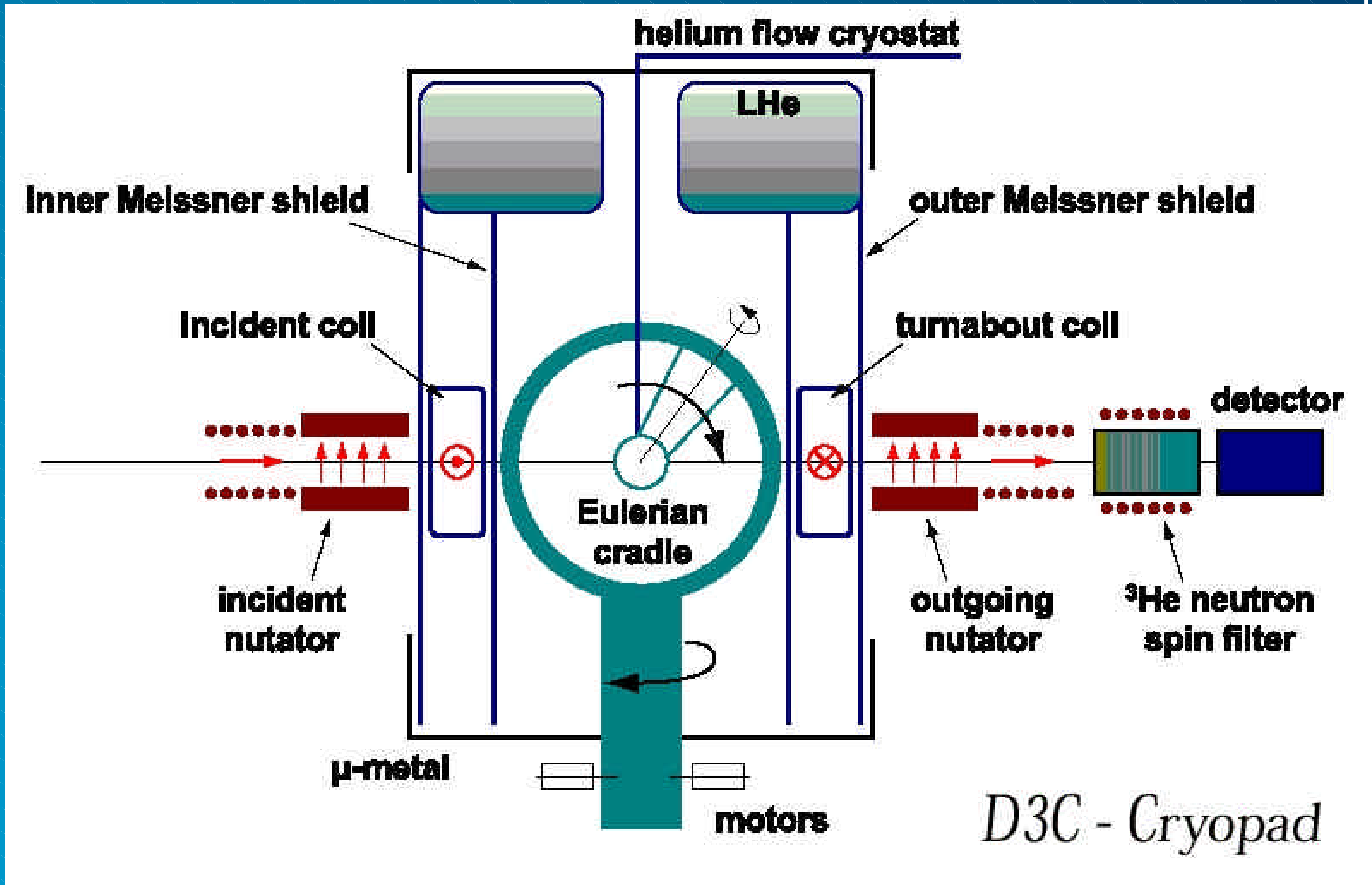
Polarized Neutrons & He3 Filters

Francis Tasset, Eddy Lelievre, Adrin Hiller, Trefor Roberts



Proposed new cryopad setup on D3C

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The Millennium Programme at ILL -> Ever Bigger Detectors

Diffraction Group

New Diffraction Group Instruments:

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- D4c – a microstrip detector for liquids & amorphous materials
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