The Millennium Programme at ILL

Alan Hewat, ILL Diffraction Group.

ILL Grenoble

Alan Hewat

Are fancy new instruments at the ILL Grenoble high flux reactor relevant for the new medium flux Australian reactor ?

Many ILL techniques have been adapted for smaller reactors:

- Cold source and neutron guides...
- Monochromators, position sensitive detectors, air pads...
- Cryostats, temperature controllers, He^3 dilution inserts...
- Small angle scattering machines, powder diffractometers...

And many ILL instruments have been adopted FROM smaller reactors

Only a factor of ~5 difference in flux –> comparable instruments

The ILL Diffraction Group

www.ill.fr/dif/

ILL Grenoble

Alan Hewat

- 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.

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The Millennium Programme at ILL
-> New Detectors

Alan Hewat

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

Alan Hewat

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)

Too many diffractometers ? (vs spectrometers)

Are we building machines that people want to use ?

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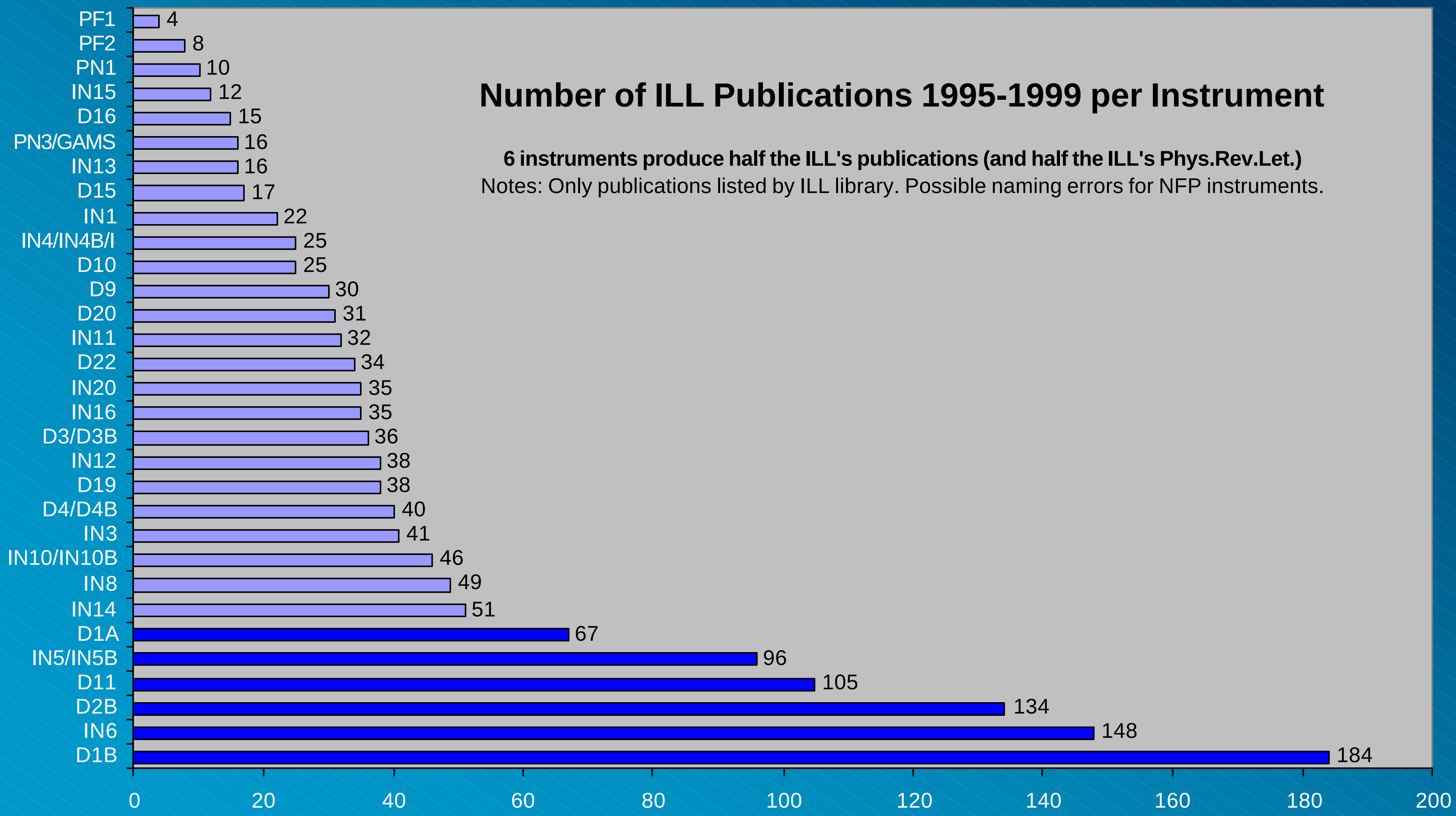

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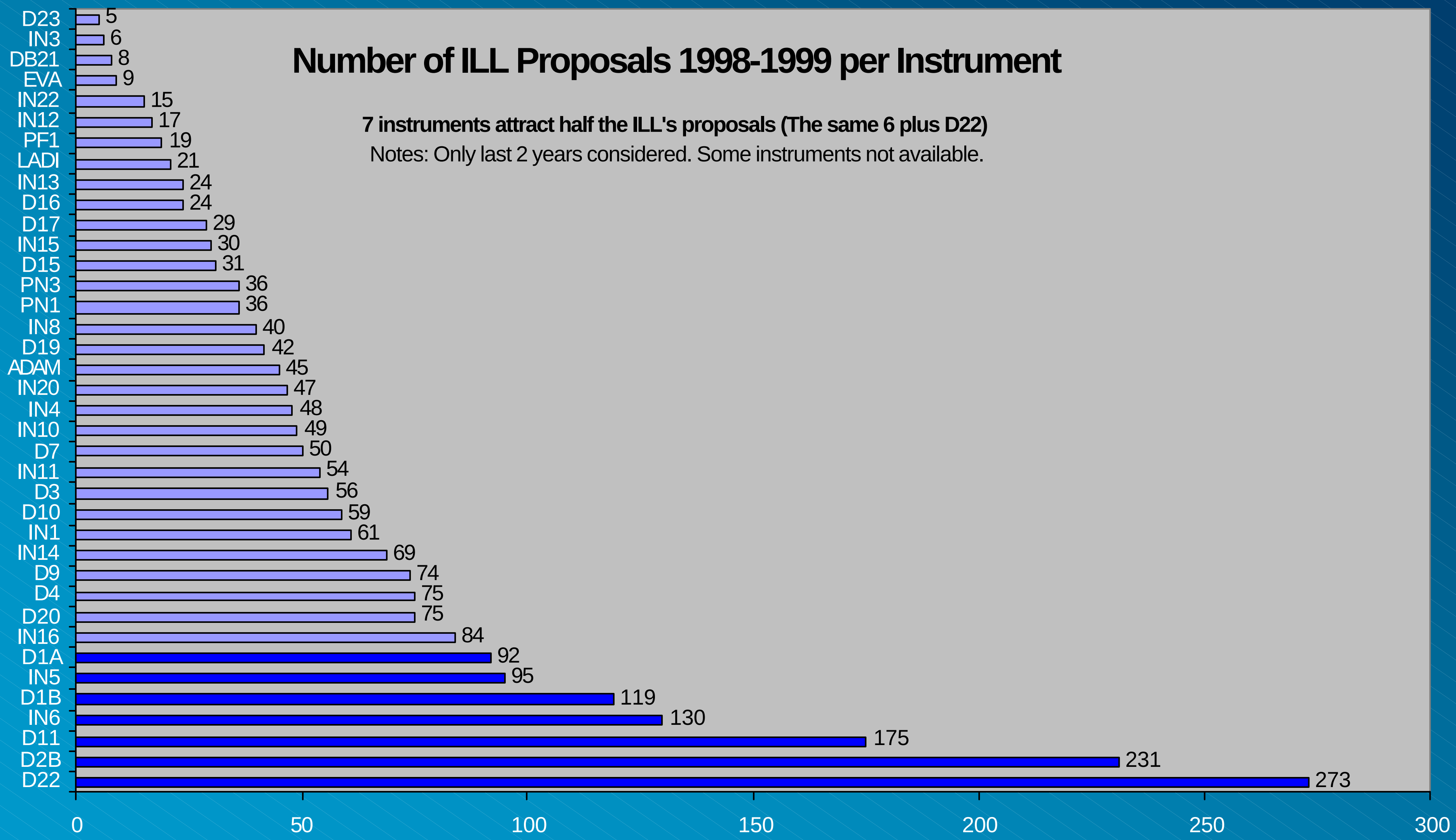
Are we building machines that people want to use ?

Alan Hewat



The ILL Millennium Programme…
-> New Detectors

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Alan Hewat

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…

What is a Microstrip Detector ?

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Instead of wires, a printed circuit is used.
This allows high resolution, mechanical stability...

Microstrip Detectors - Printed Circuits

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Alan Hewat

"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)

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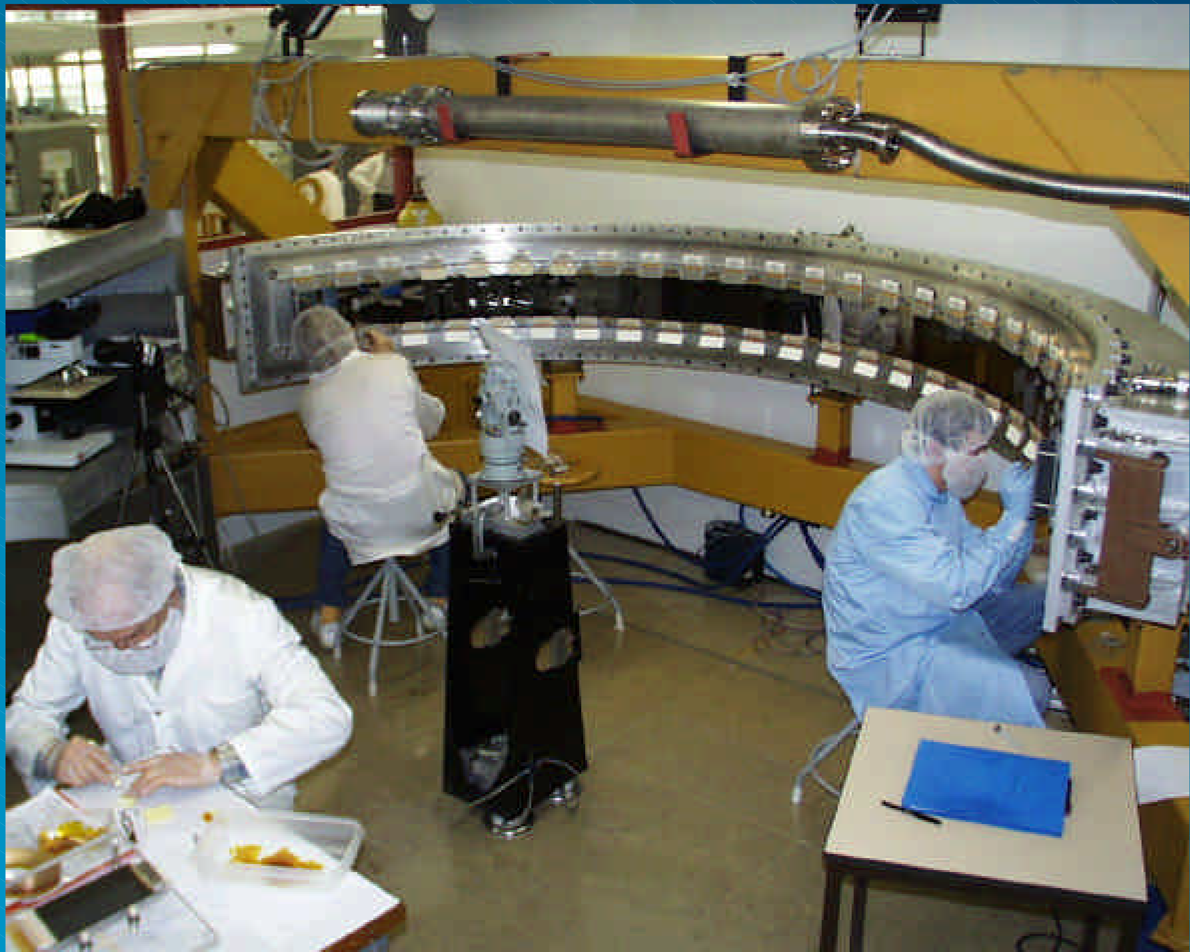
Alan Hewat

The 160º D20 Microstrip Array



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

High Flux Powder Diffractometer D20

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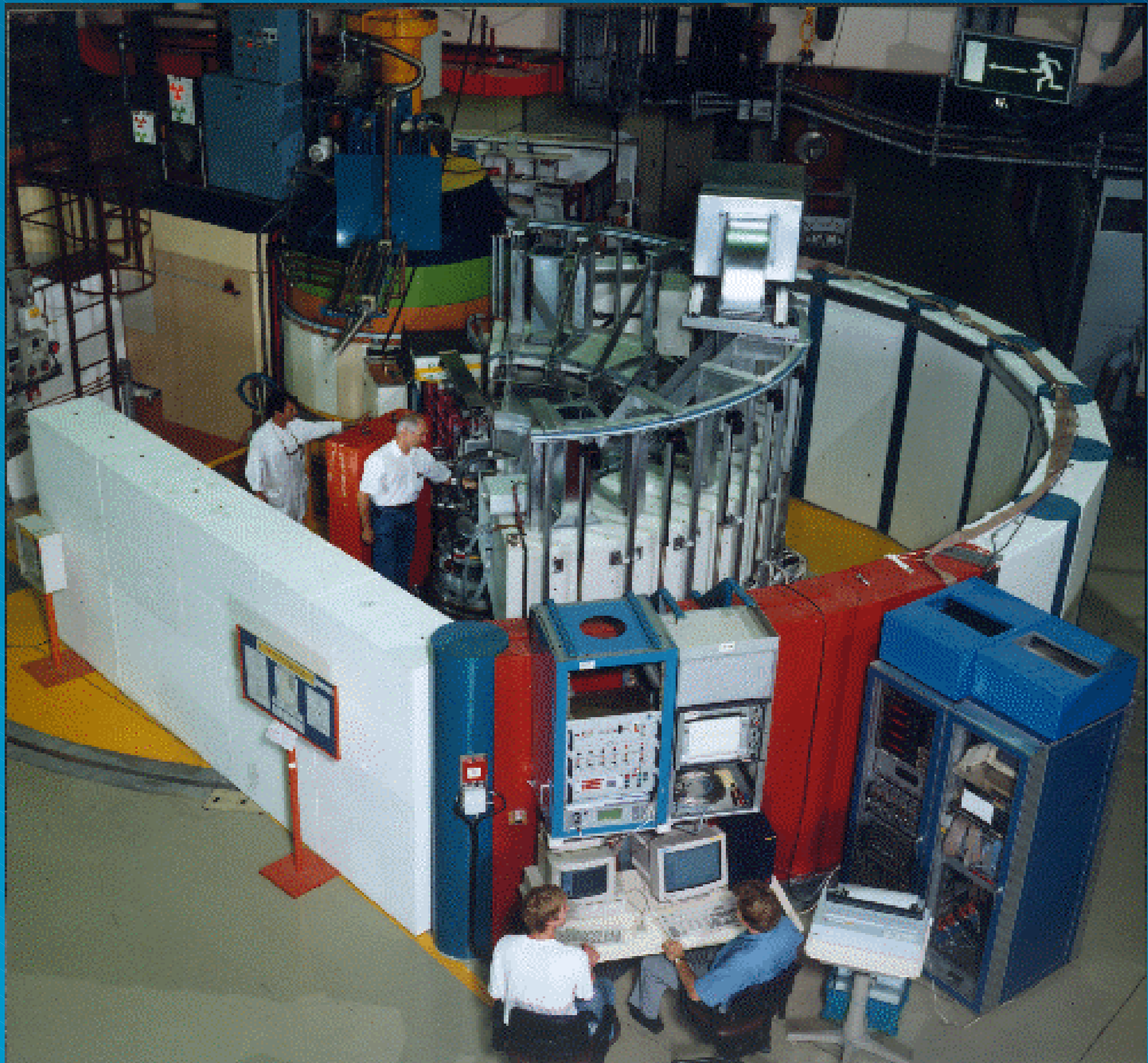
Pierre Convert, Thomas Hansen, Jacques Torregrossa



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

ILL Grenoble

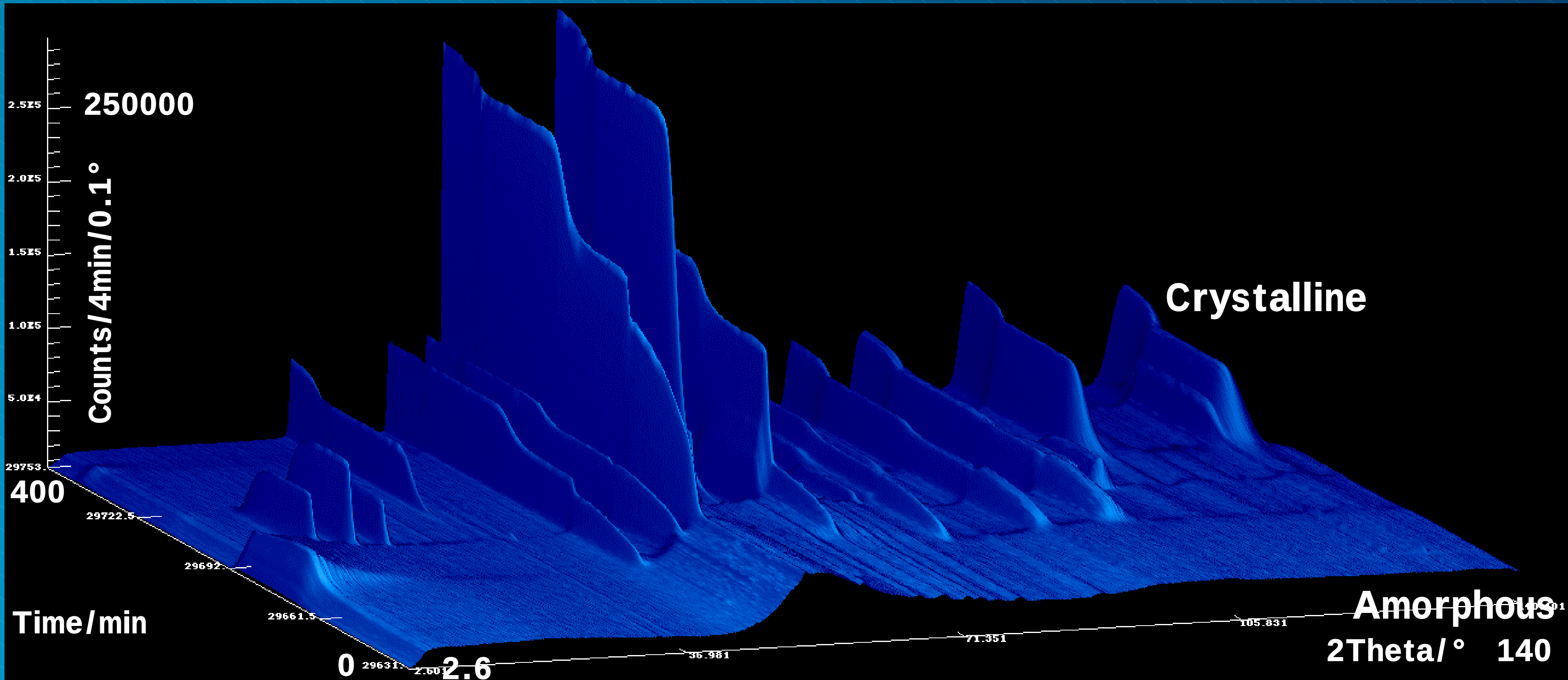
Alan Hewat

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

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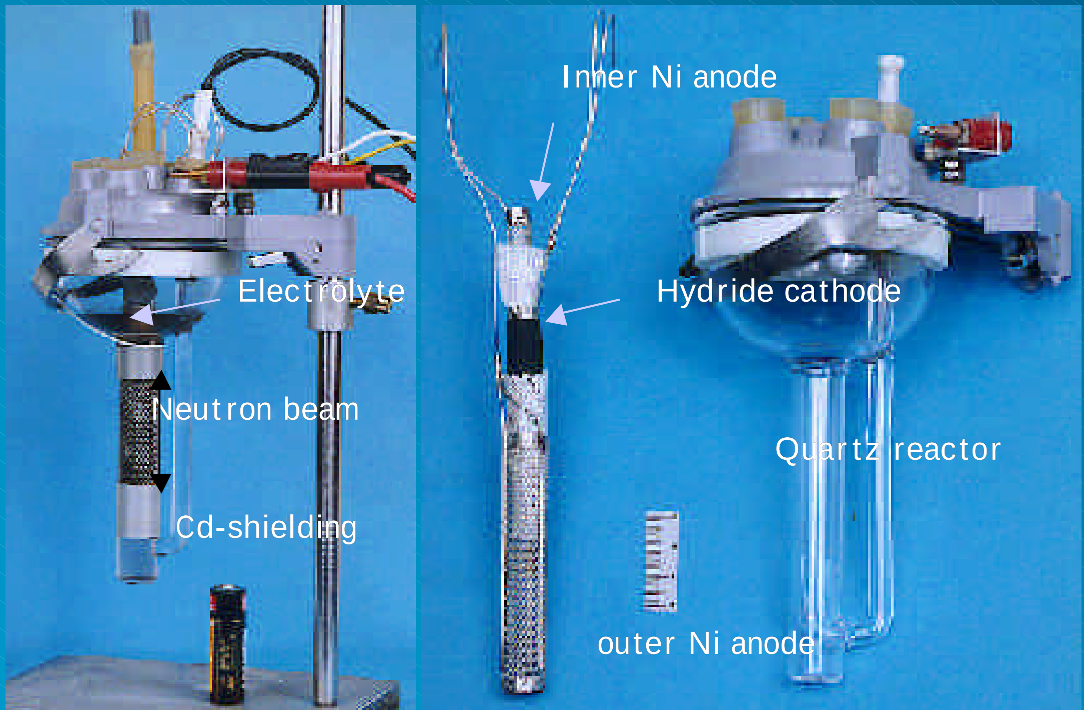
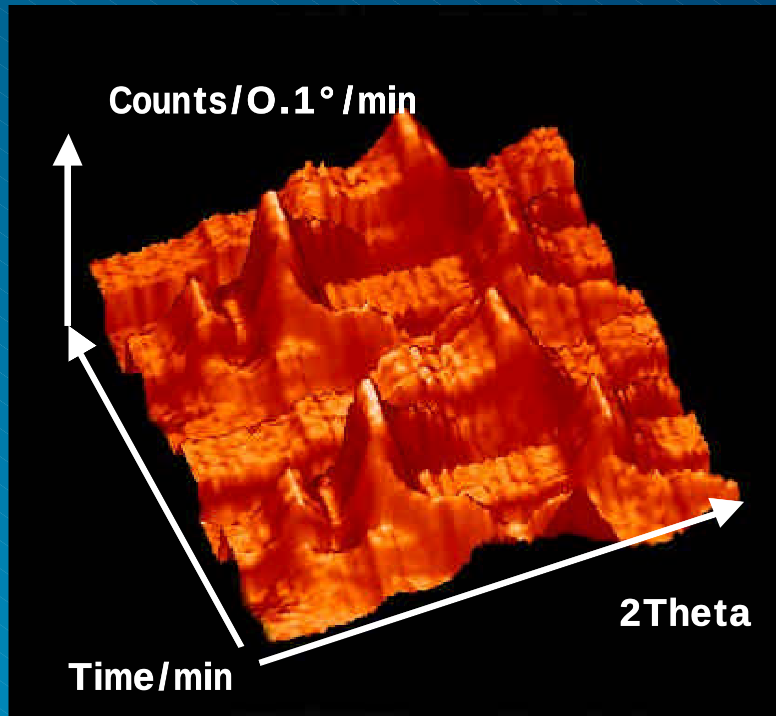
Applications of large fast detectors
Real-time electro-chemistry

Alan Hewat

Latroche, Chabre et al.:

In-situ Charging and discharging of metal hydride electrodes LaNi5





- Follow chemical changes with battery charge/discharge cycle

Applications of large fast detectors
Real-world samples

Alan Hewat

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

High Pressure Microstrip Detectors

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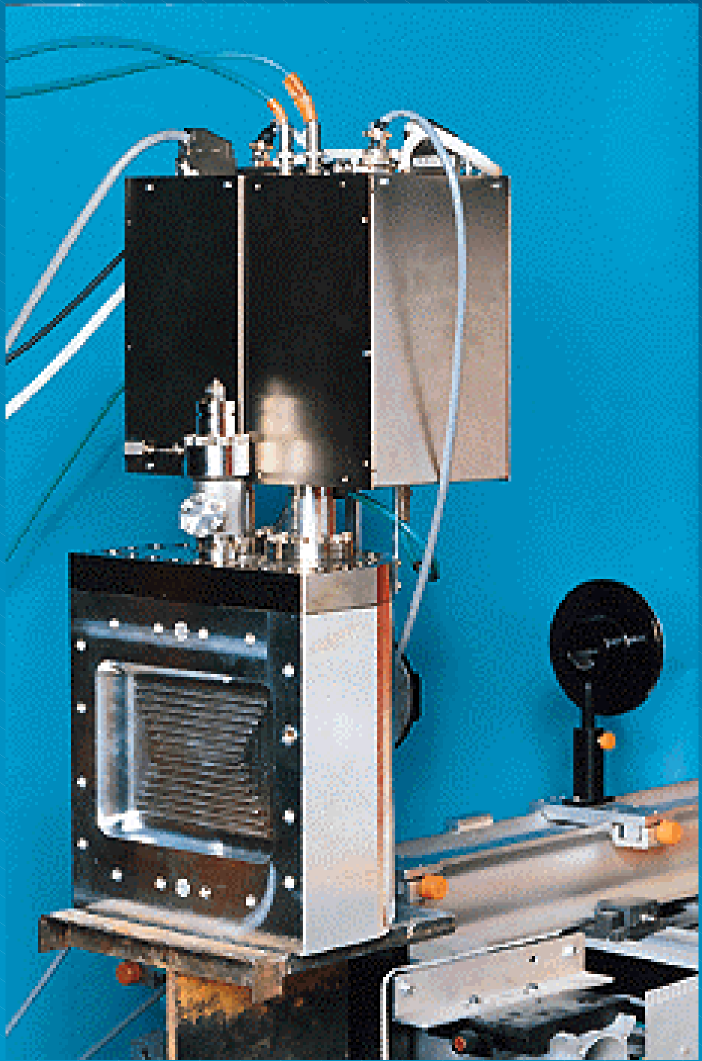
New D4C Liquids & Amorphous Materials Diffractometer

Henry Fischer, Gabriel Cuello, Pierre Palleau

Alan Hewat



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

The prototype D4C detector

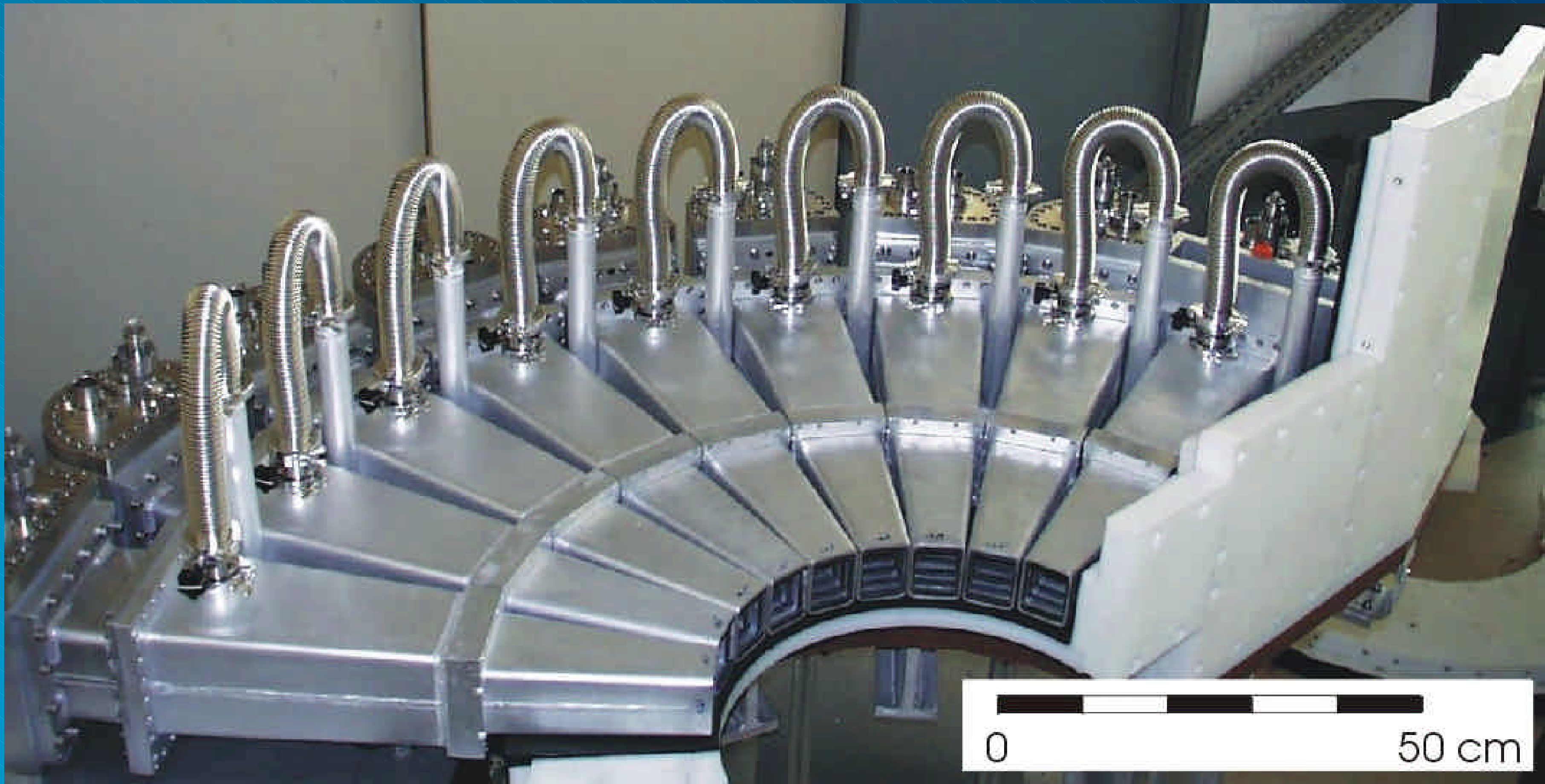
An array of Microstrip Detectors

New D4C Liquids & Amorphous Materials Diffractometer

Henry Fischer, Gabriel Cuello, Pierre Palleau

ILL Grenoble

Alan Hewat



Very high efficiency & stability needed for isotope replacement method

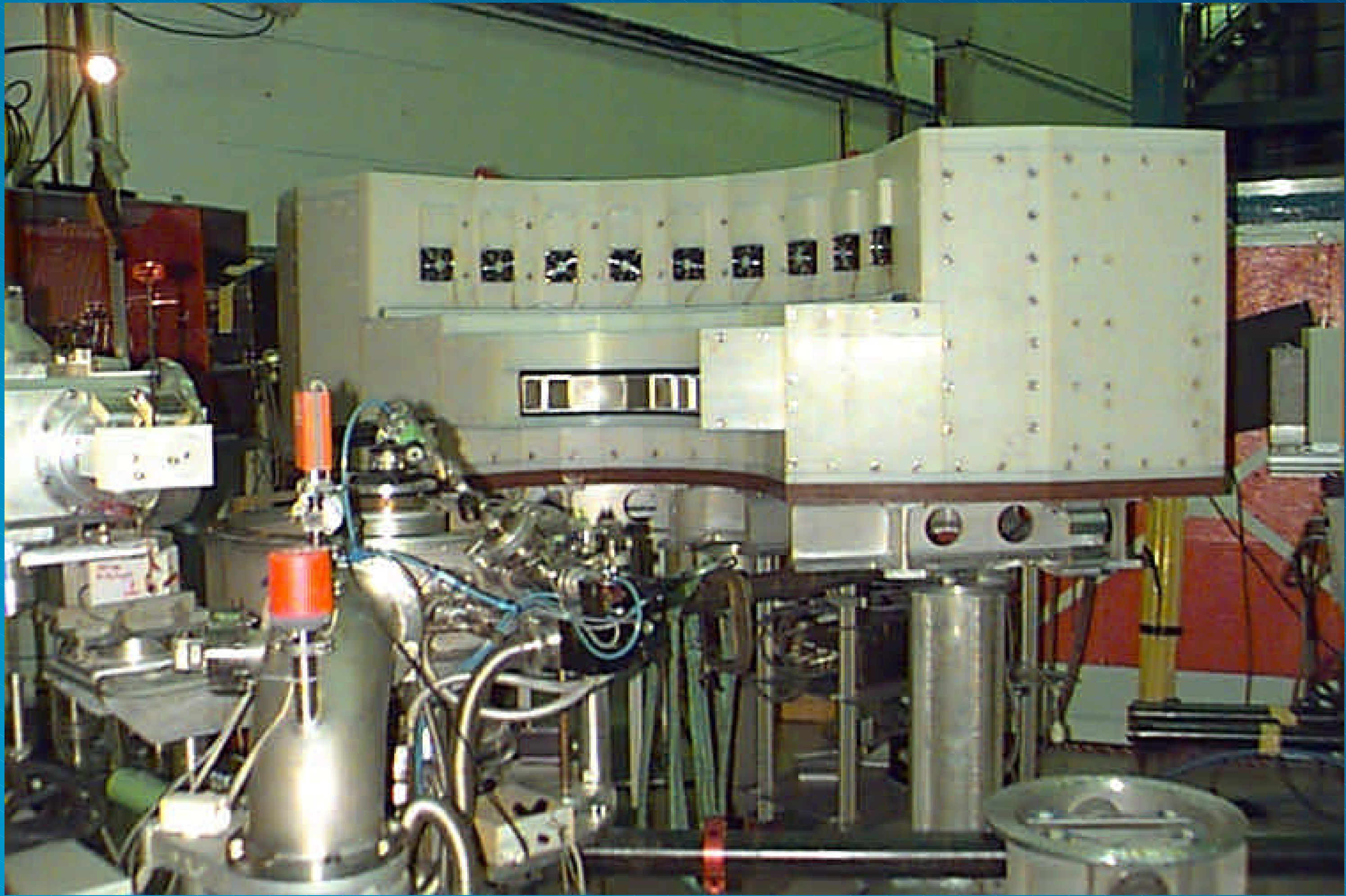
An array of Microstrip Detectors

ILL Grenoble

New D4C Liquids & Amorphous Materials Diffractometer

H.E. Fischer, P. Palleau, D. Feltin (2000) Physica B 276-278, 93

Alan Hewat

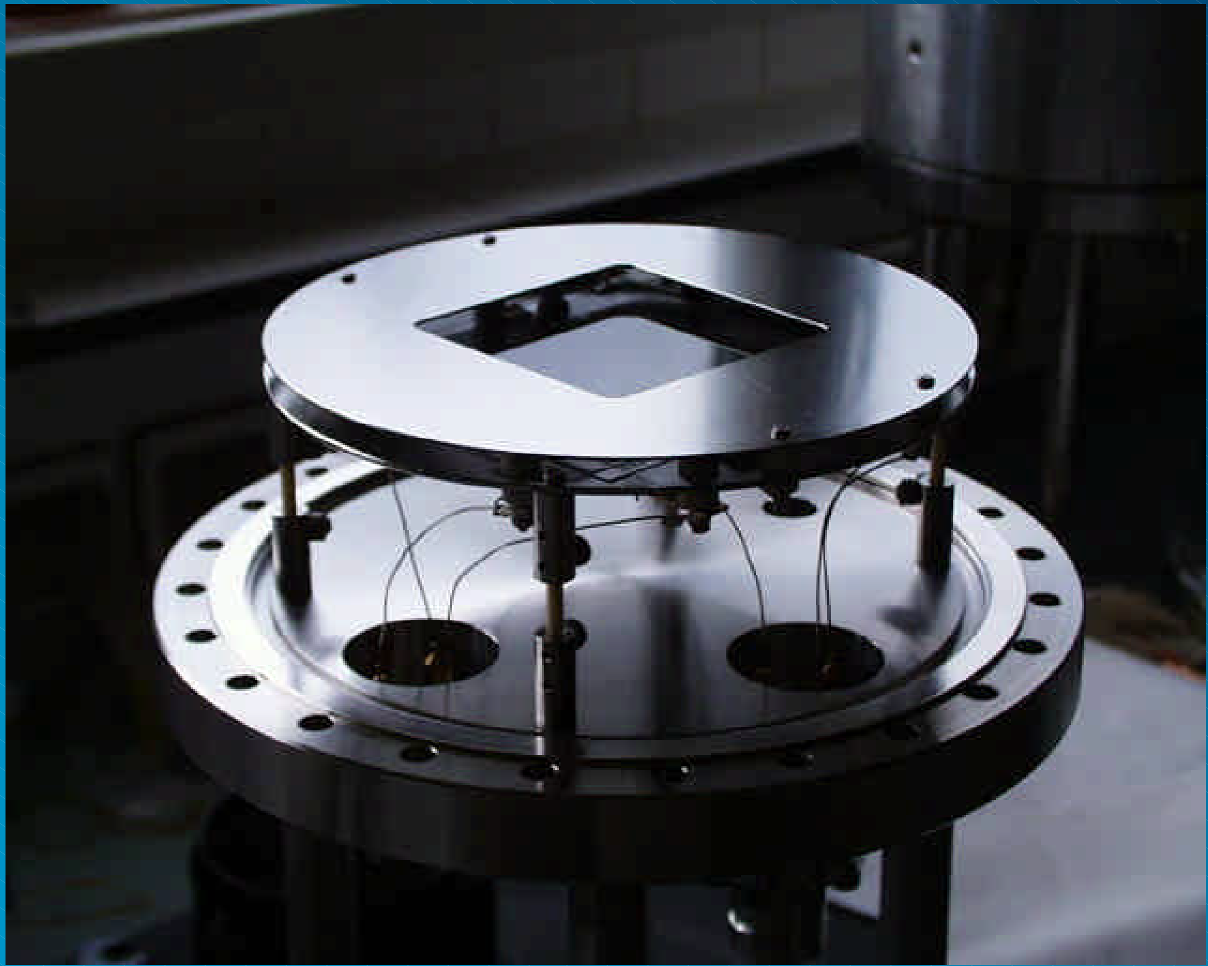


New D4C in place with shielding (June 2000)

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

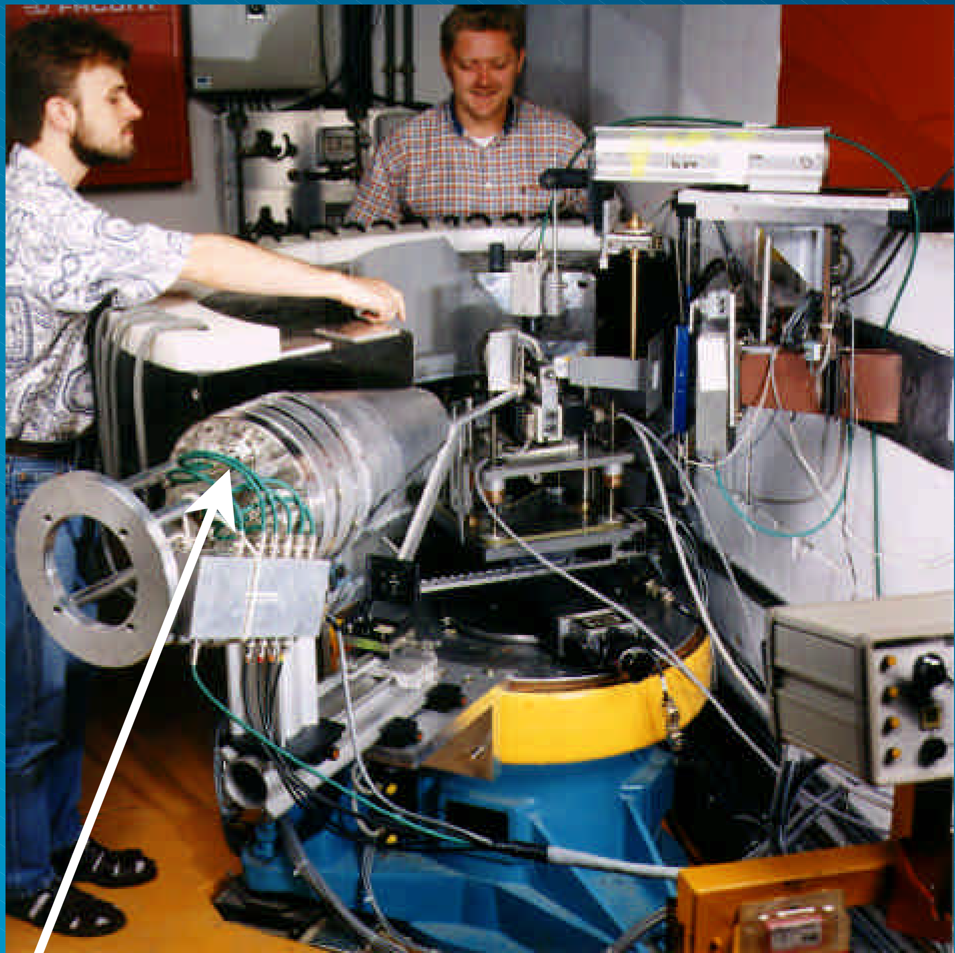
Neutron Strain Scanner

ILL Grenoble

80x80 mm 2D Microstrip Detector on D1A

Thilo Pirling, Robert Wimpory

Alan Hewat

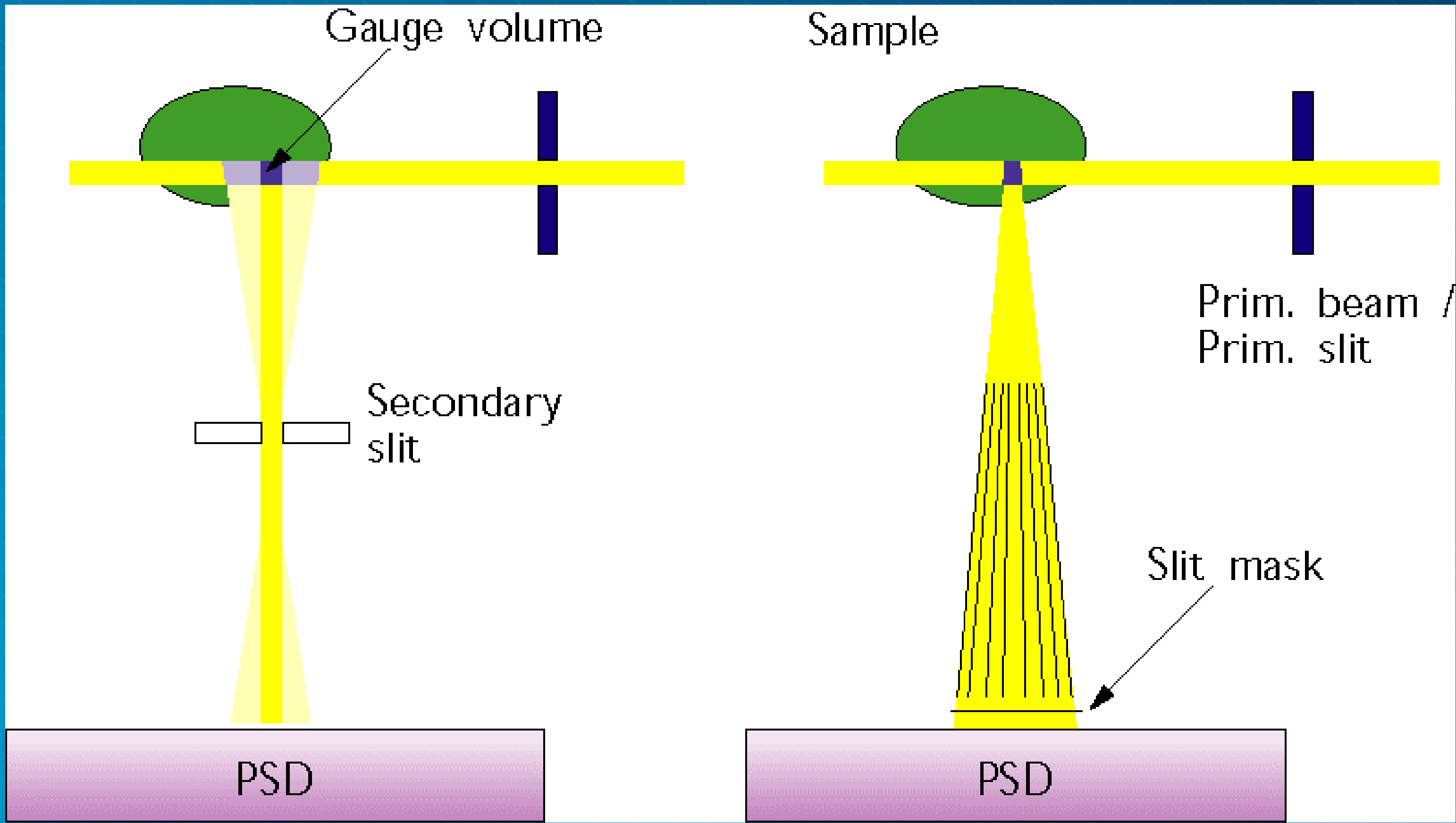


The 2D microstrip detector is used to obtain the complete line profile all at once

Neutron Strain Scanner

80x80 mm 2D Microstrip Detector on D1A

Thilo Pirling, Robert Wimpory



A large convergent collimator is used with the 2D microstrip detector

Note the very small sampling volume with this setup (right)

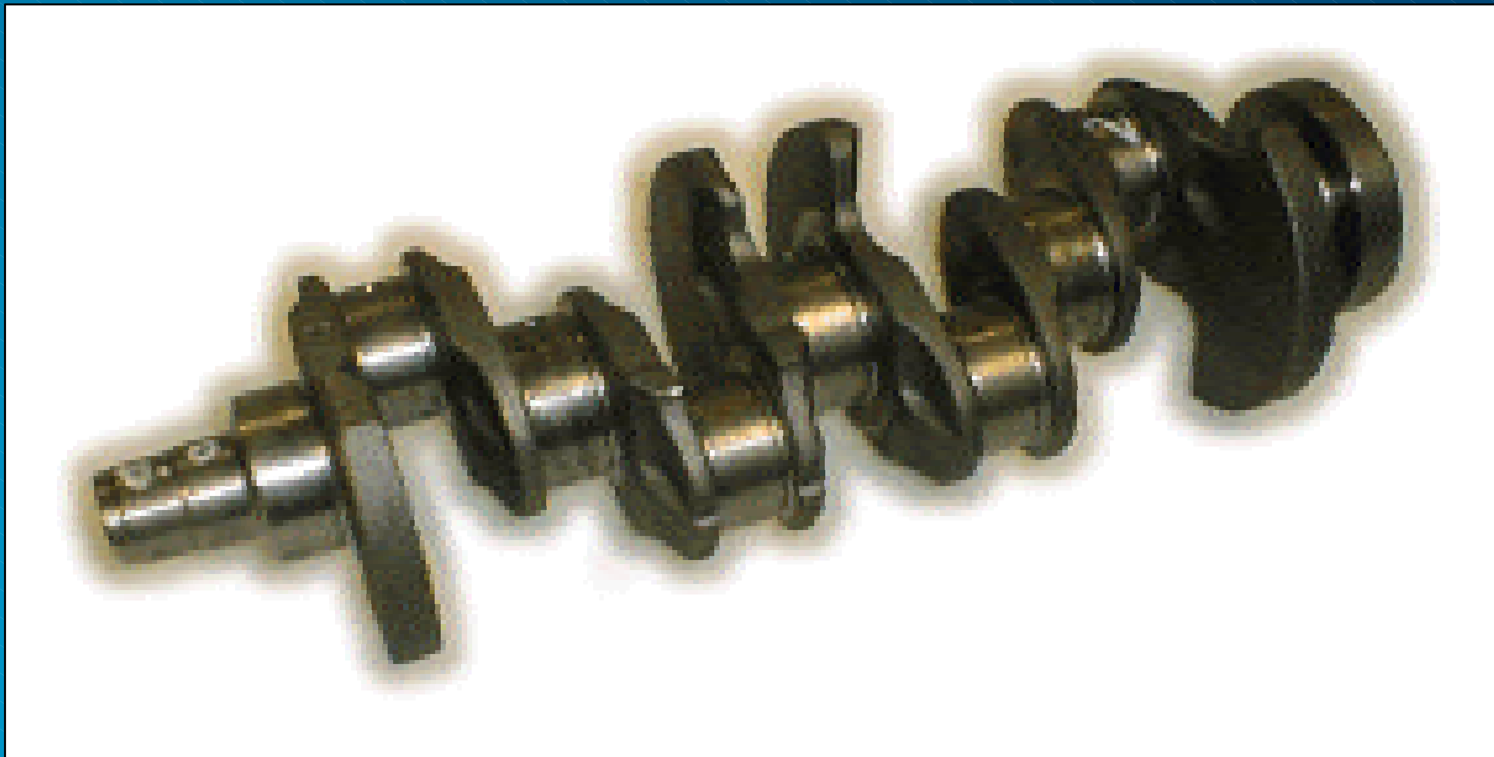
Neutron Strain Scanner

80x80 mm 2D Microstrip Detector on D1A

Thilo Pirling, Robert Wimpory

ILL Grenoble

Alan Hewat



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

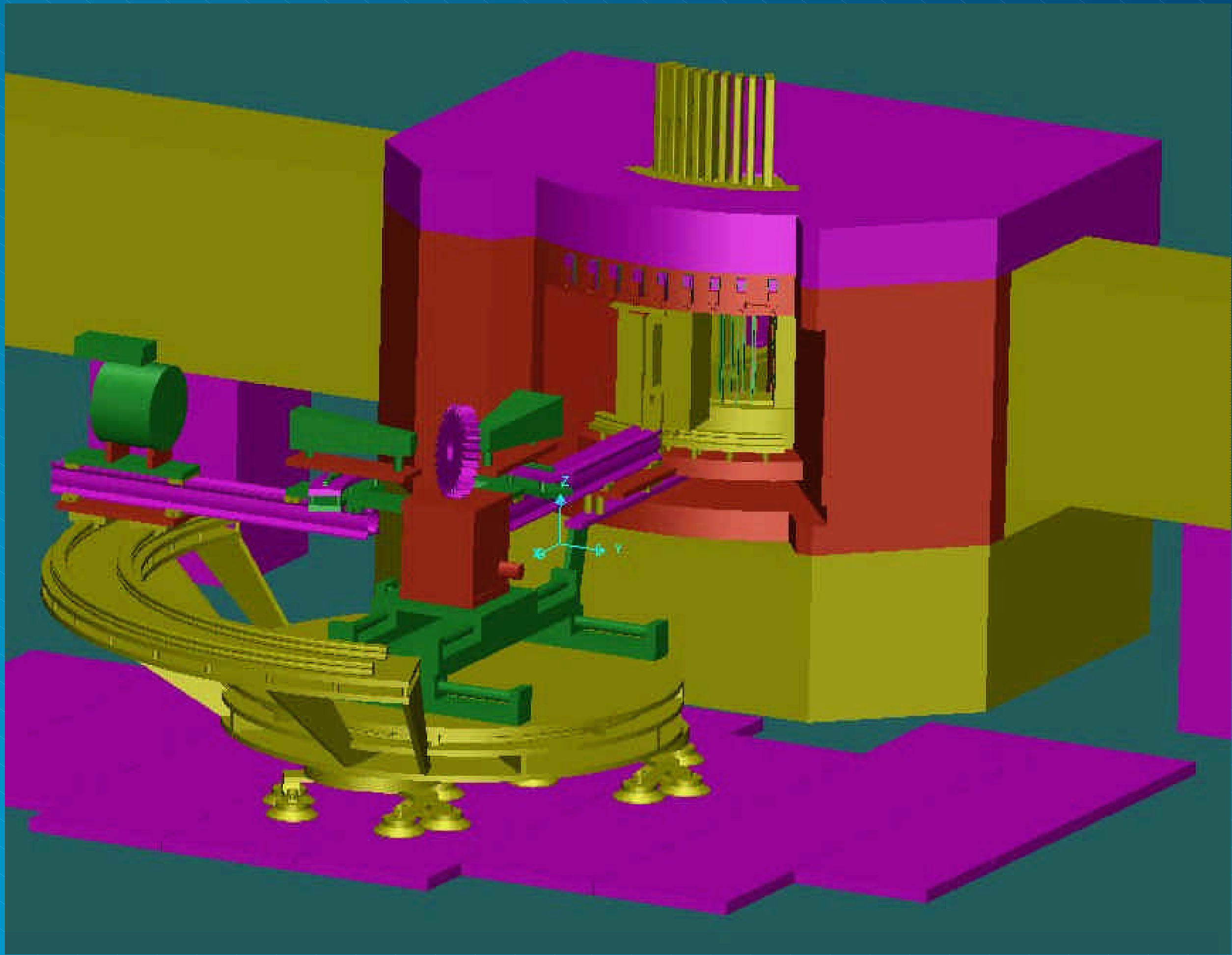
A New ILL-EPSRC Strain Scanner

EPSRC grant of ~ 1M Pounds Sterling

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

ILL Grenoble

Alan Hewat



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

ILL Grenoble

Alan Hewat

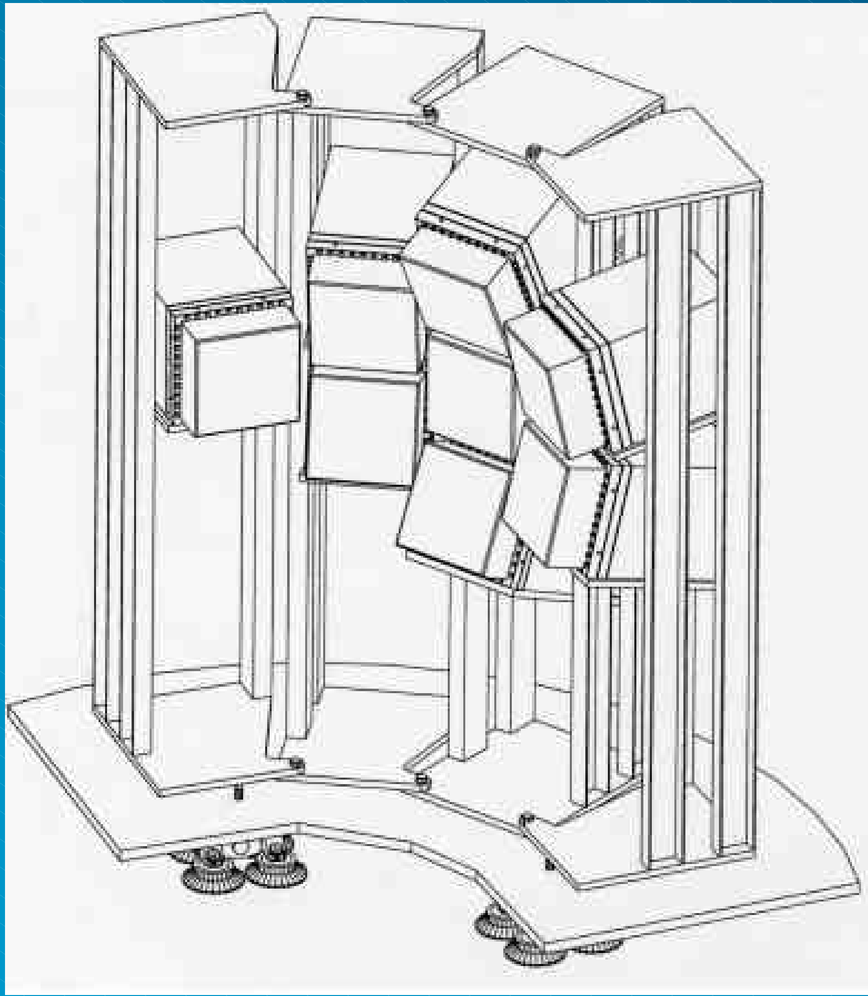
200x200 mm 2D microstrip detector for D19 fibre & protein diffractometer

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

ILL Grenoble

Sax Mason, Trevor Forsyth, John Archer, Michael Walsh

Alan Hewat



9 Independent 2D microstrip detectors

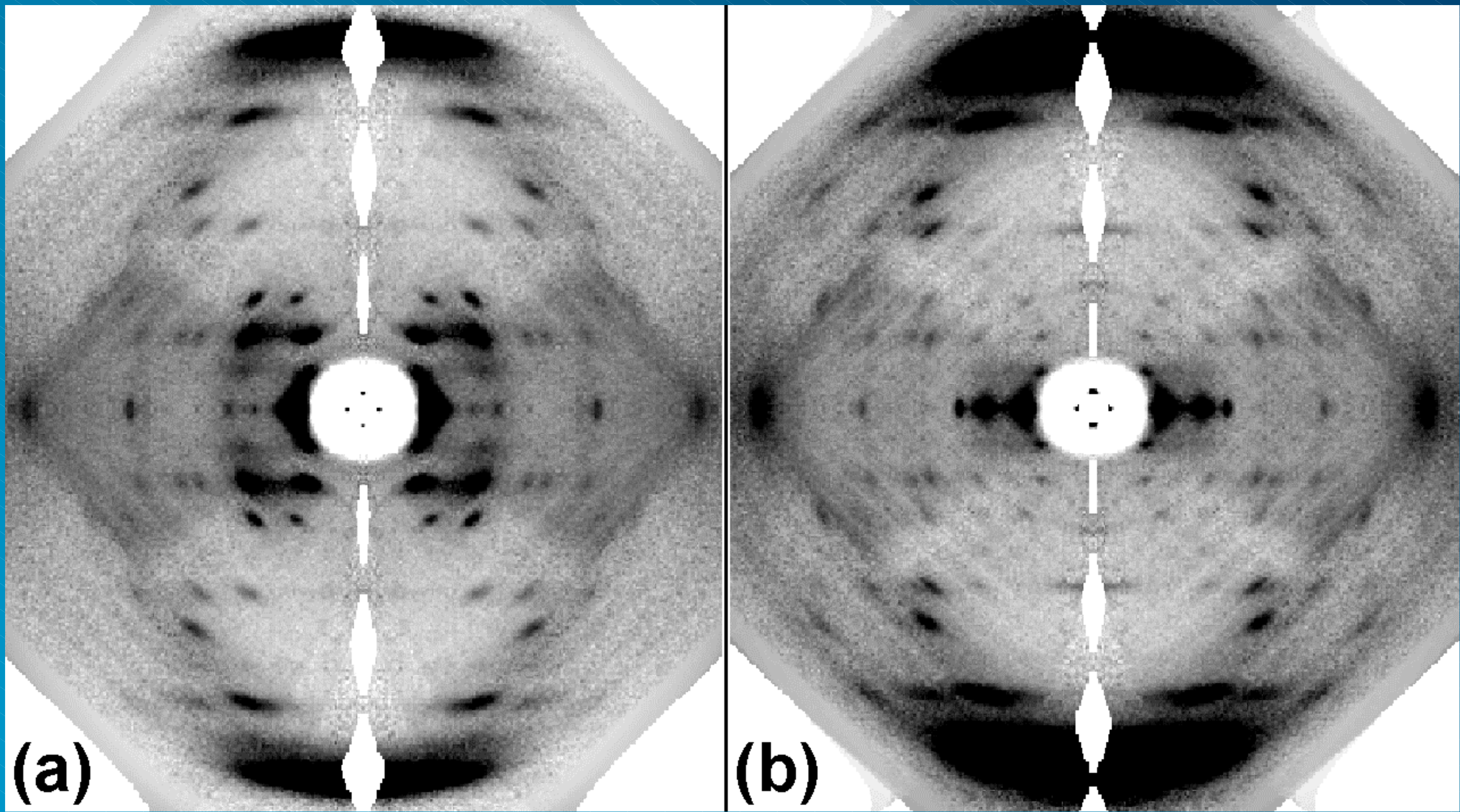
- 15 year old D19 detector covers only a thin 2D strip
- Replace with an array of high resolution 2D modules
- Increase efficiency x20
- Fibre Diffraction
  Small protein structures
  In-situ hydration studies.

Water in B-DNA sheets on D19

ILL Grenoble

Alan Hewat

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.

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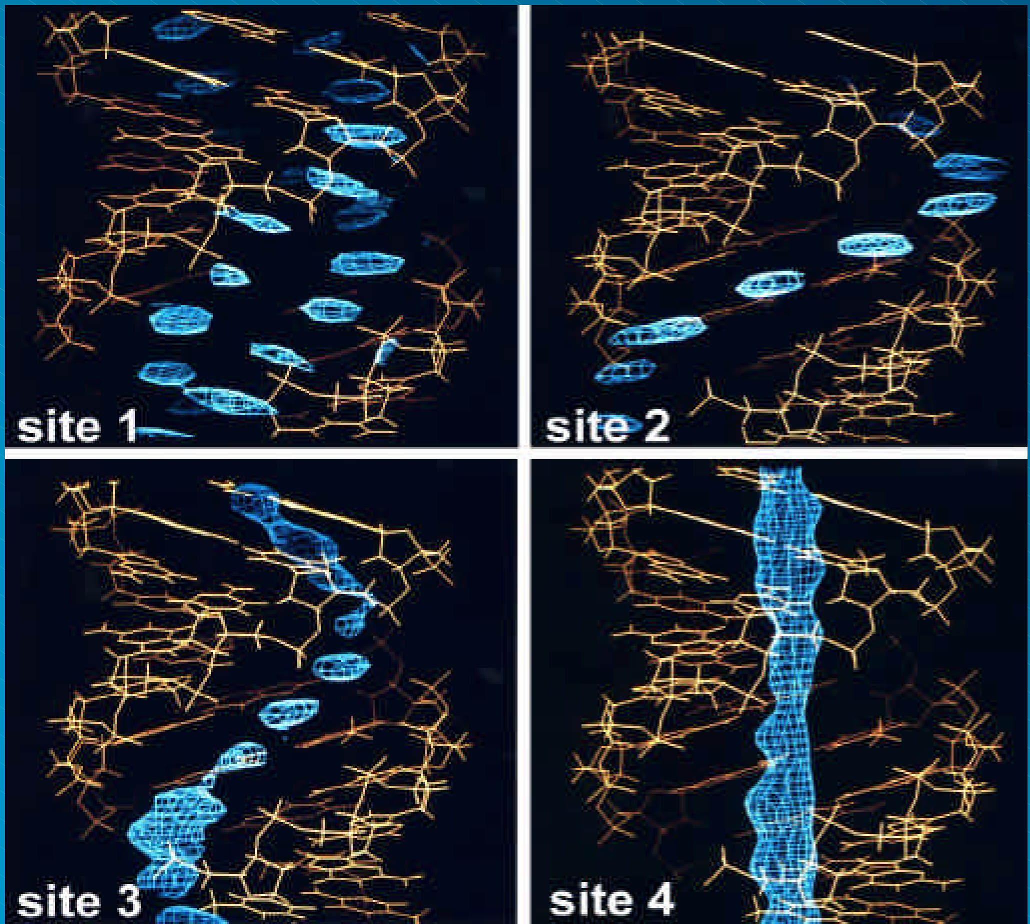

- B-DNA sheets, but A-DNA fibres
- 100 individual DNA fibres in D_2O
- Diffuse fibre diffraction patterns from D19 used to locate water
- 4 distinct water sites located along double helix backbone

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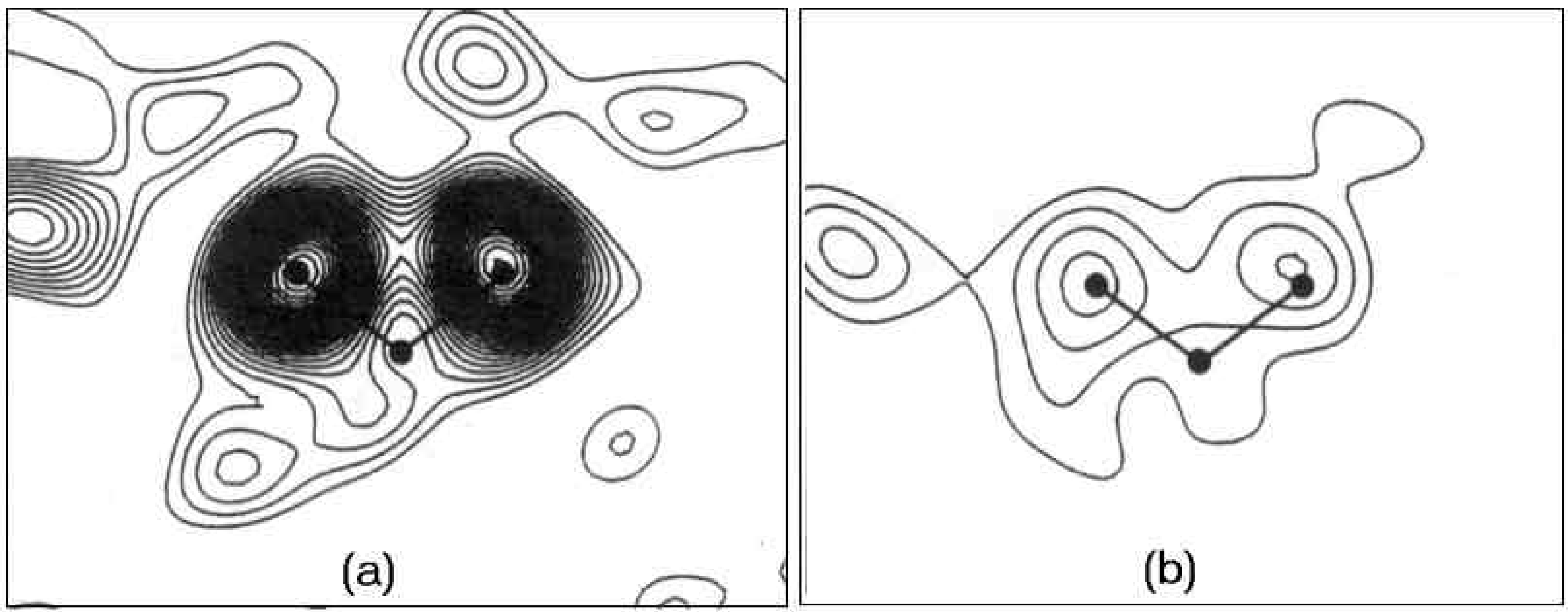
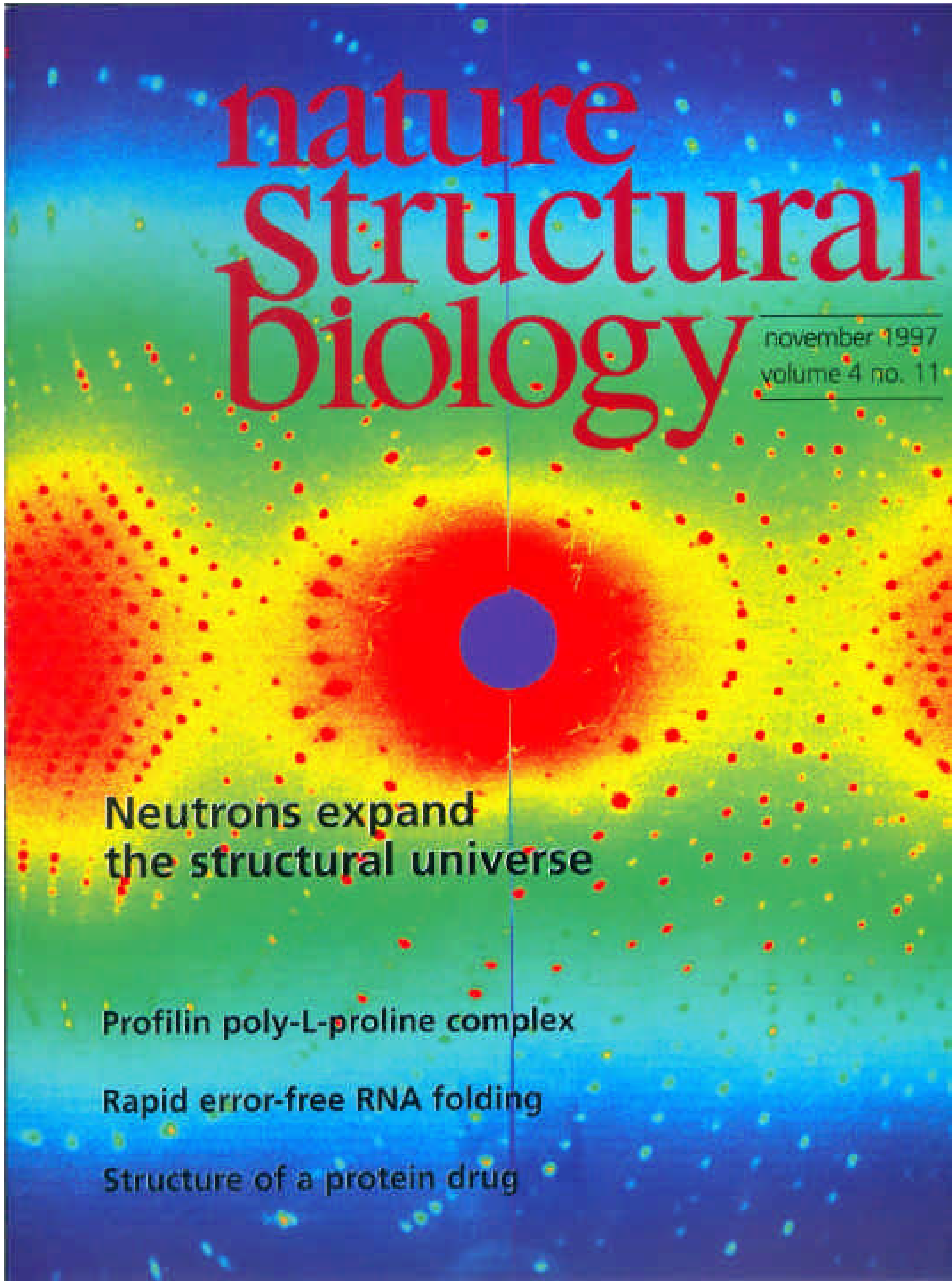
Langan, Lehmann, Wilkinson, Jogl, Kratky (1999) Acta Cryst D55, 51



D19 Neutron data

Synchrotron data



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Neutron Image Plates & Microstrip Detectors

Nature (1997) Cover showing LADI data

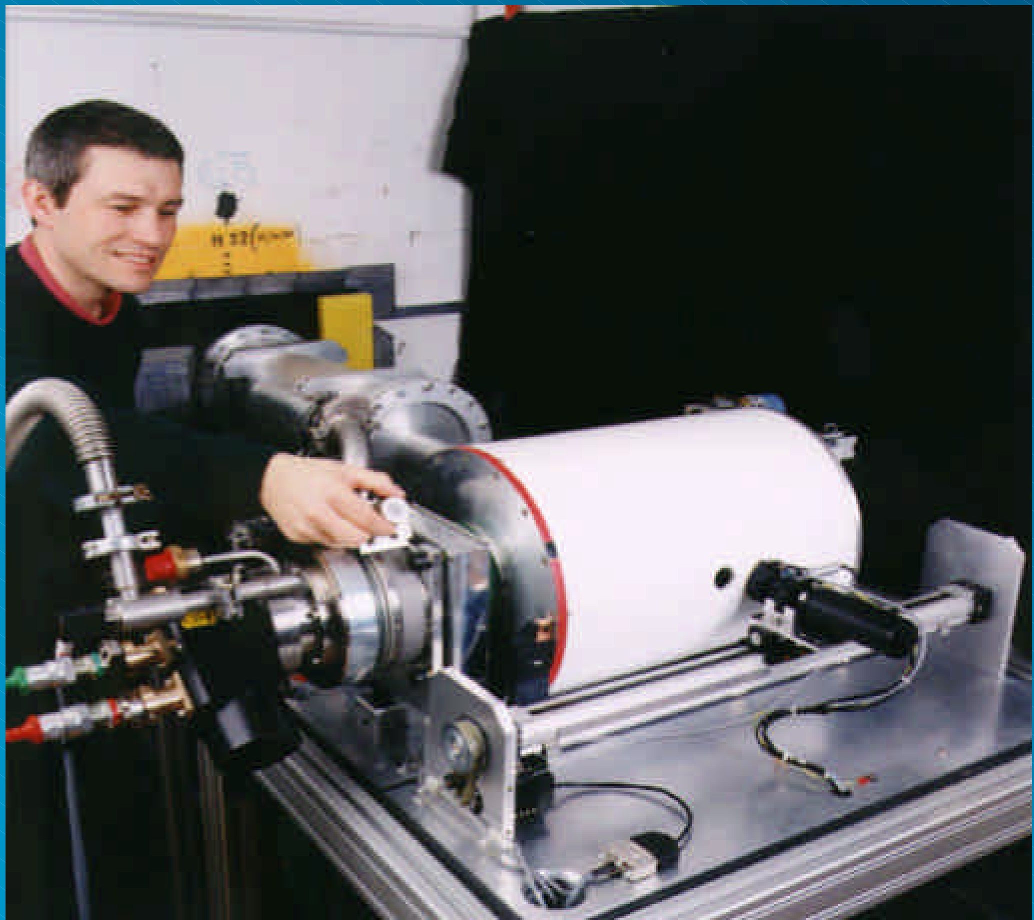
(LAue Diffractometer with Image plates)

T-LADI Laue Neutron Image Plate for physics and chemistry

Dean Myles, Clive Wilkinson, Garry McIntyre

ILL Grenoble

Alan Hewat



- 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

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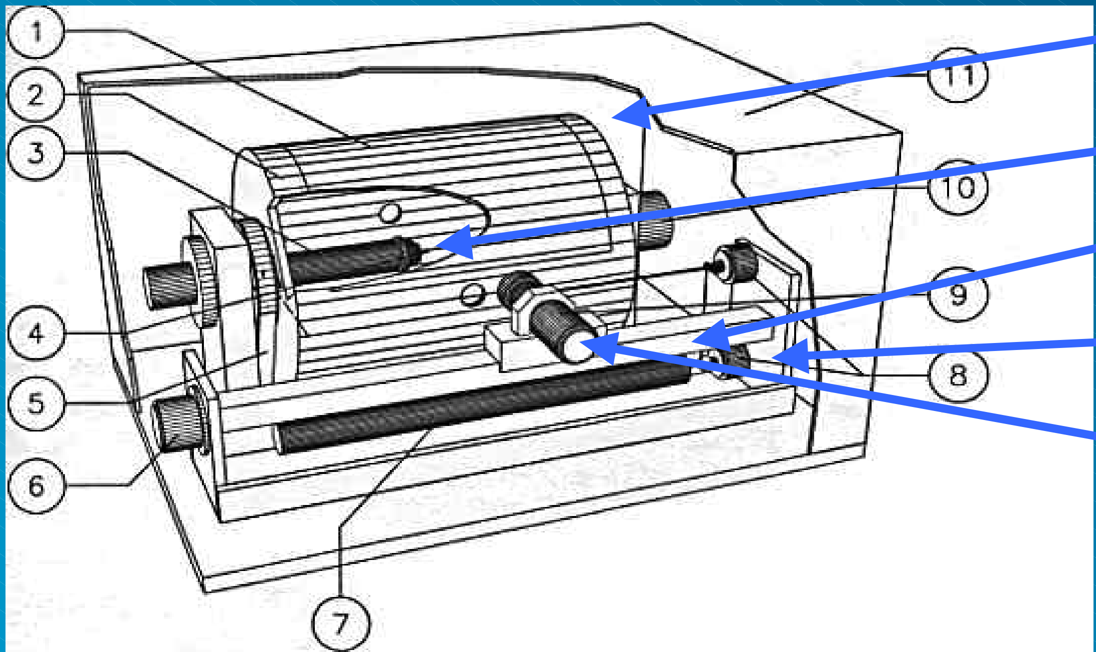
T-LADI Neutron Image Plate for physics and chemistry

Dean Myles, Clive Wilkinson, Garry McIntyre

Alan Hewat



1. Image plate on rotating drum

3. Sample holder

7. He-Ne laser

8. Focussing mirrors

9. Reader head, photomultiplier

Phonograph readout time 4 min.

4000x2000 pixels of 200 mm

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

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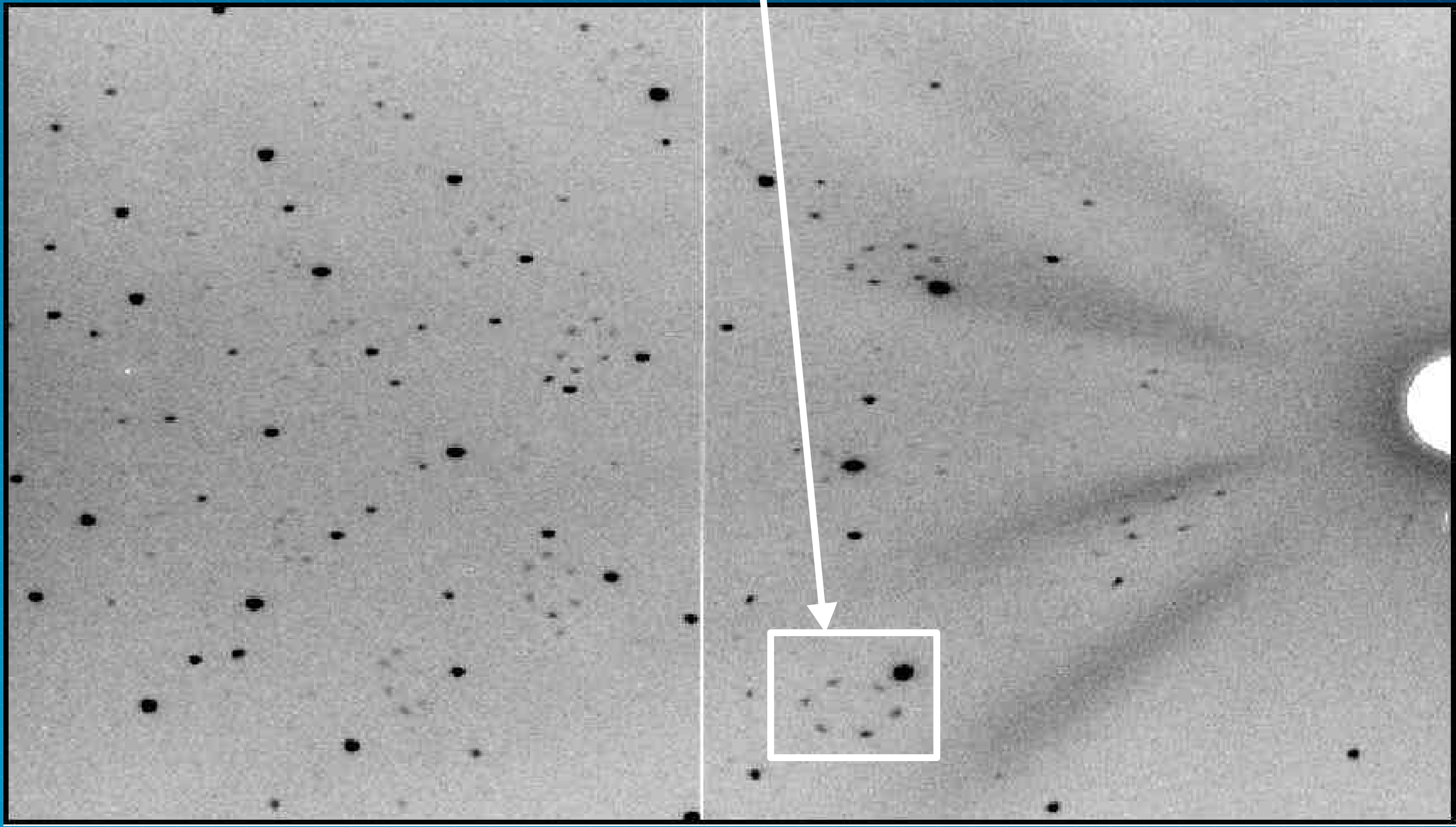
T-LADI Neutron Image Plate
Superstructure in $La_2Co_{1.7}$

Alan Hewat



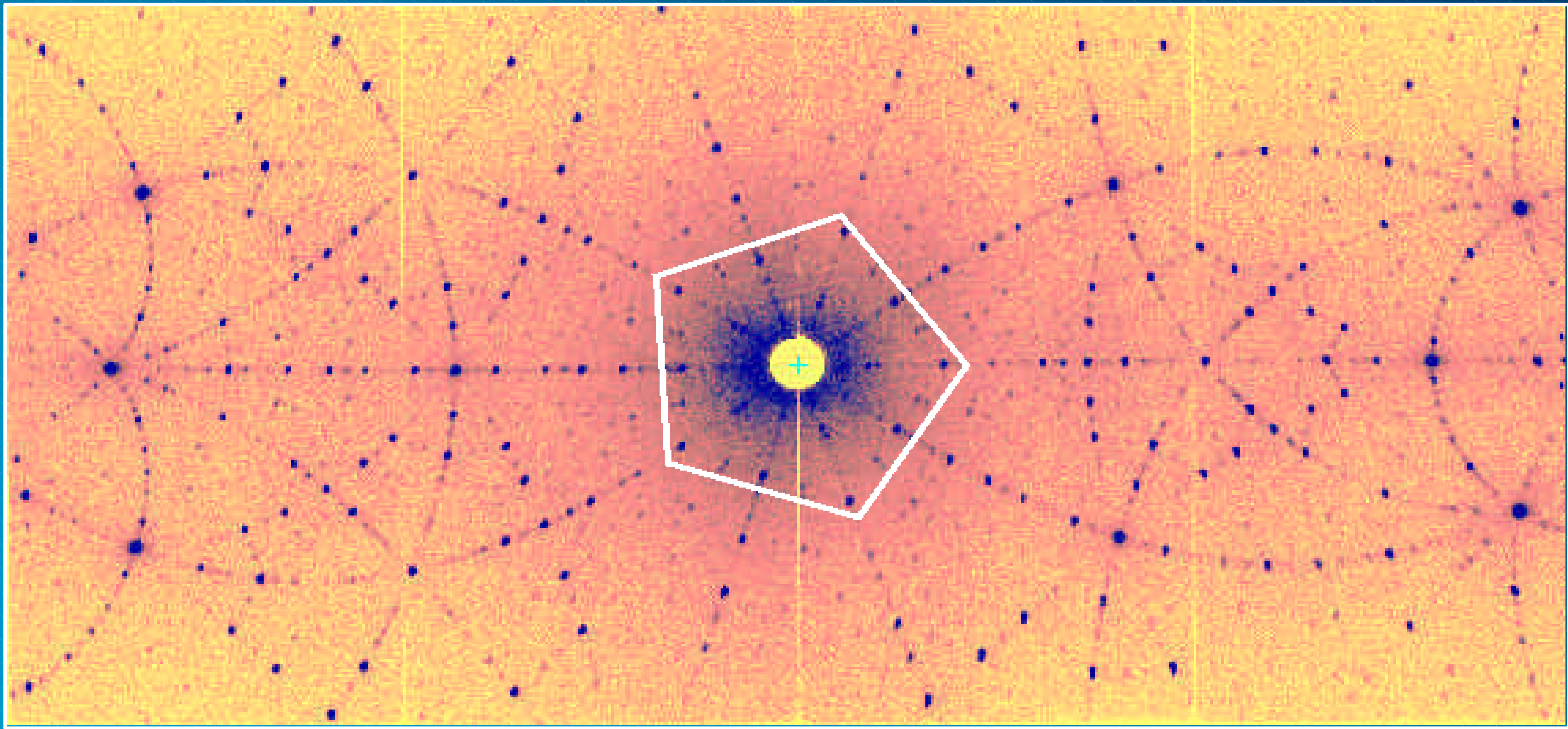
$La_2Co_{1.7}$ on T-LADI showing incommensurable superstructure

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

ILL Grenoble

Alan Hewat

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

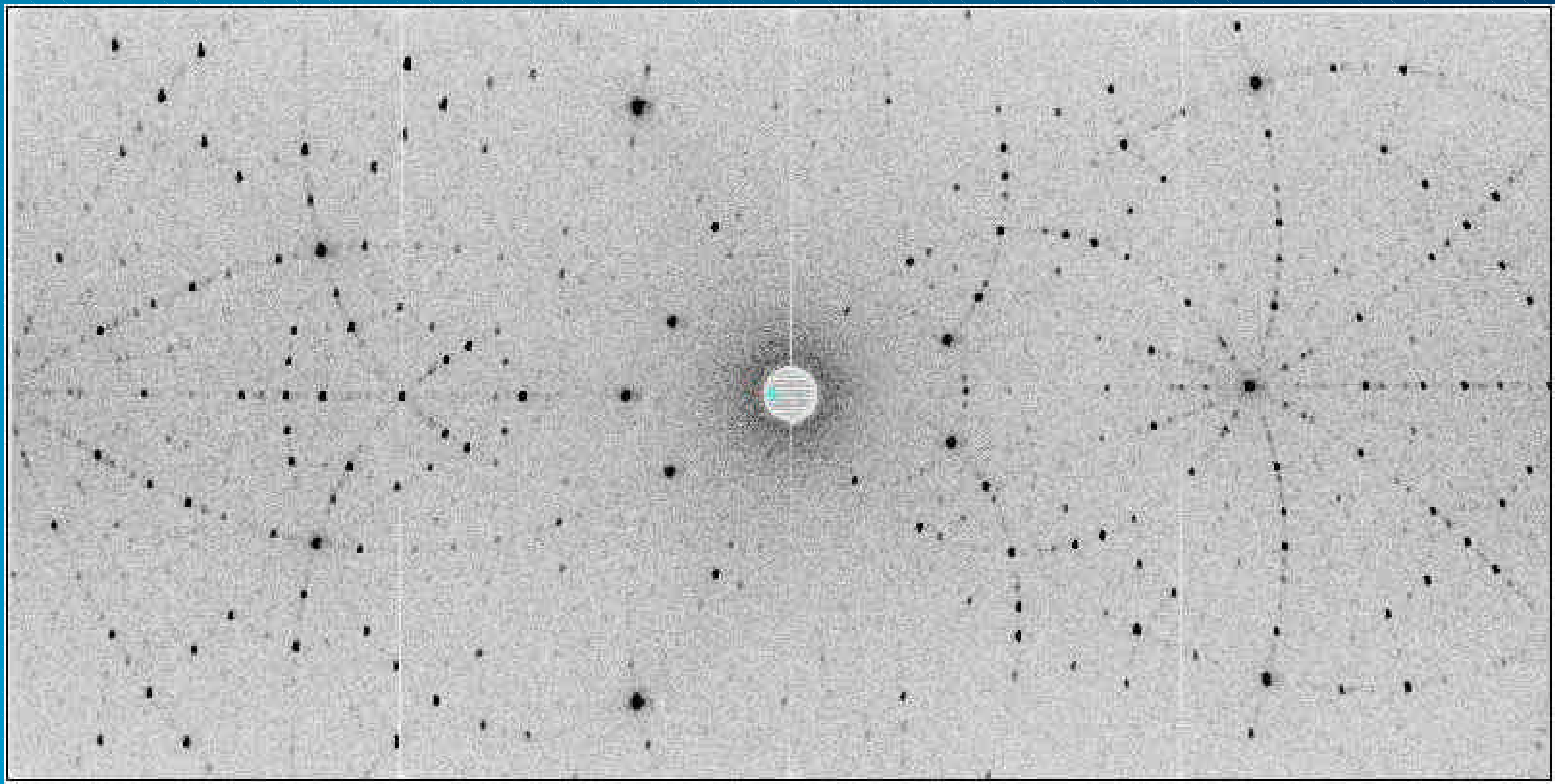


T-LADI neutron image plate photo courtesy of G. McIntyre, Oct 1999

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

ILL Grenoble
Alan Hewat

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



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

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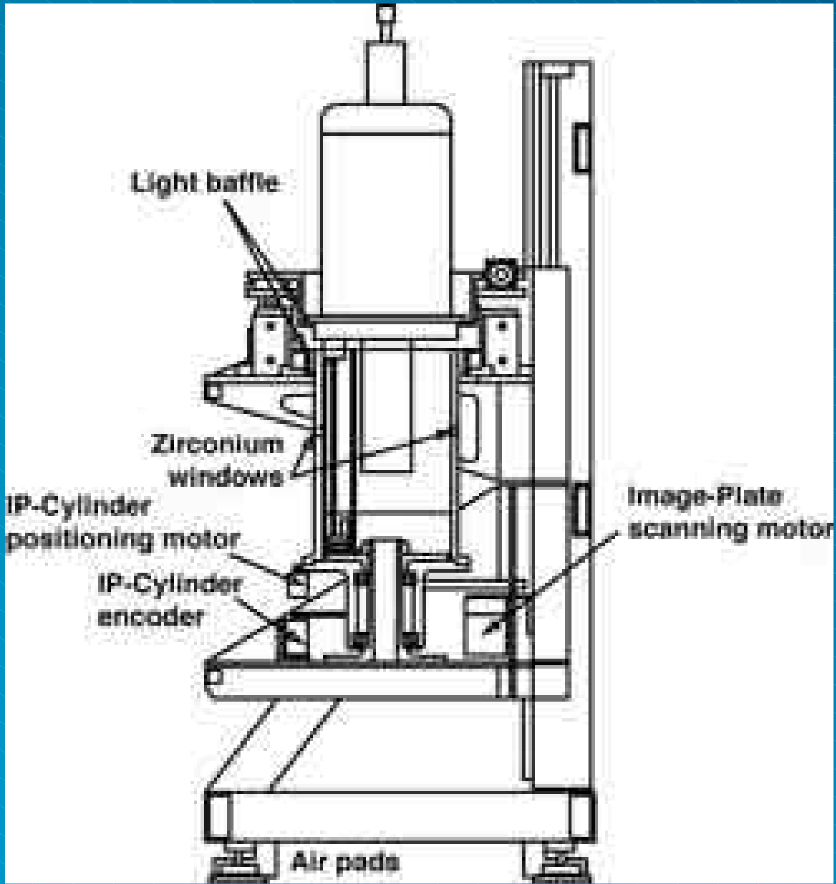


Disadvantages of Image-plates

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

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

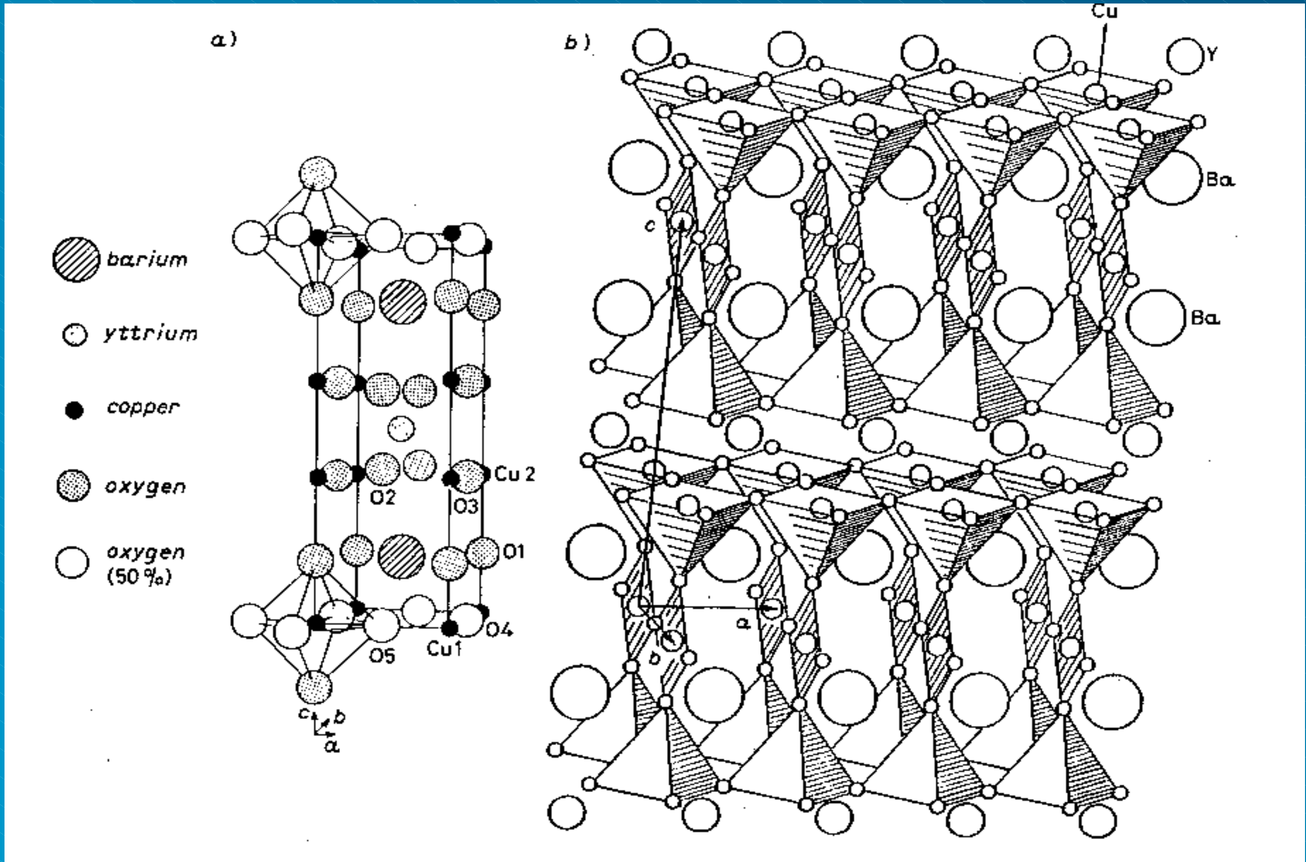
New T-LADI uses thermal neutrons, more efficient interior read-out optics, vertical geometry allowing use of cryostats, furnaces, magnets, pressure cells

ILL Grenoble

T-LADI for small crystals comparable impact to neutron powder diffraction

Alan Hewat

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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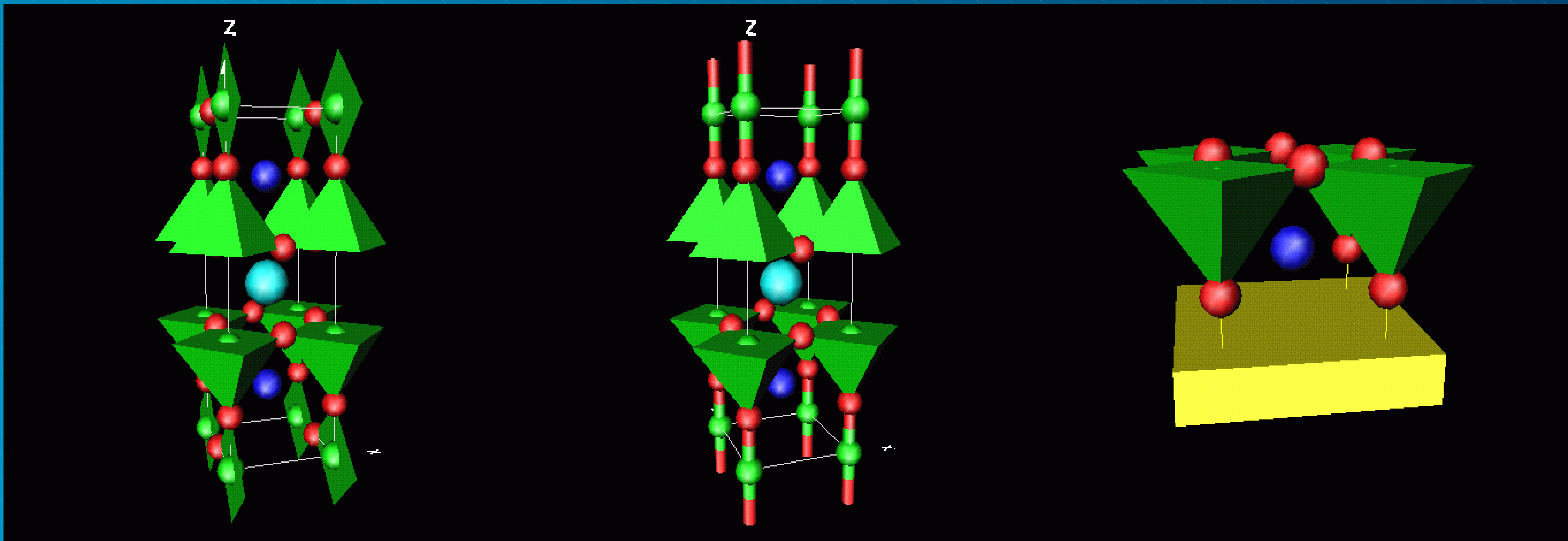
Neutron Powder Diffraction
Essential technique for new materials

Alan Hewat

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)

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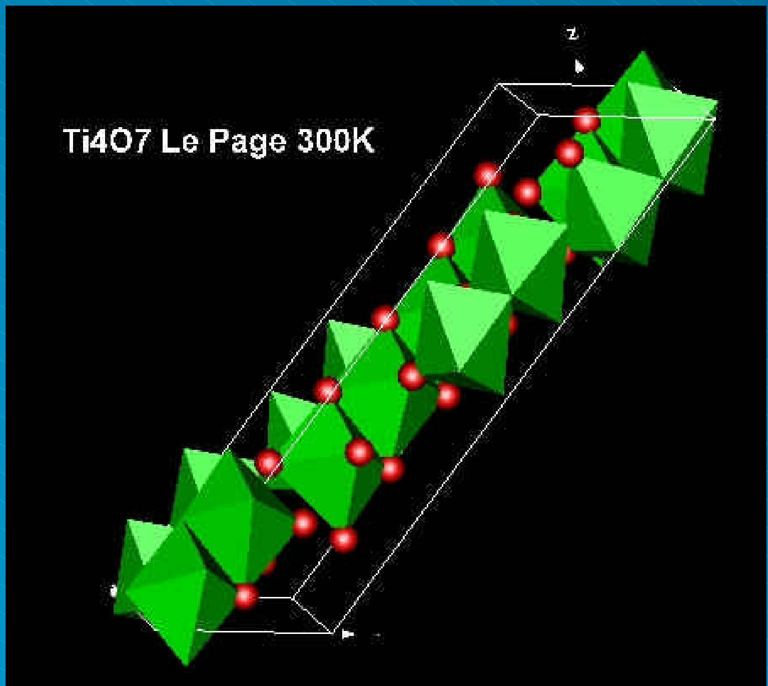
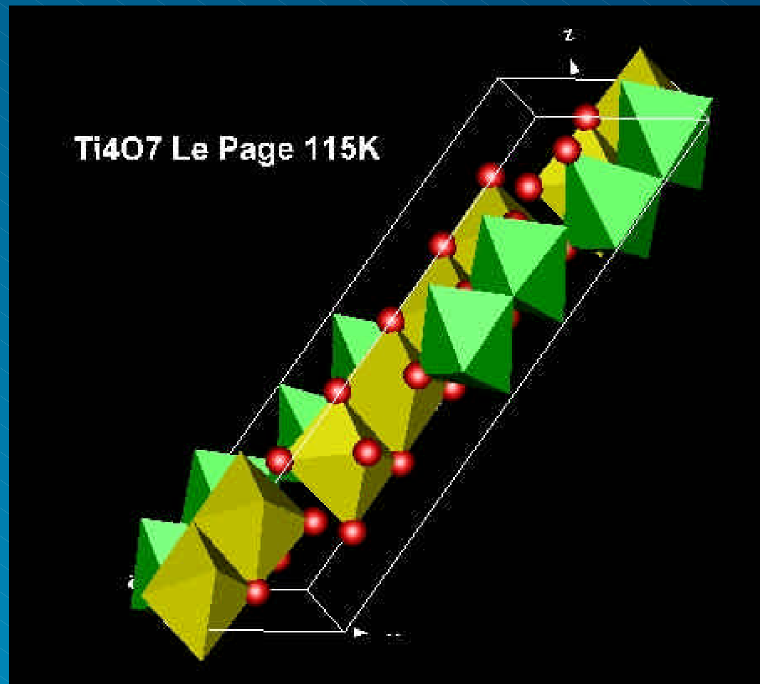
Electronic Order-Disorder

- **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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Neutron Powder Diffraction

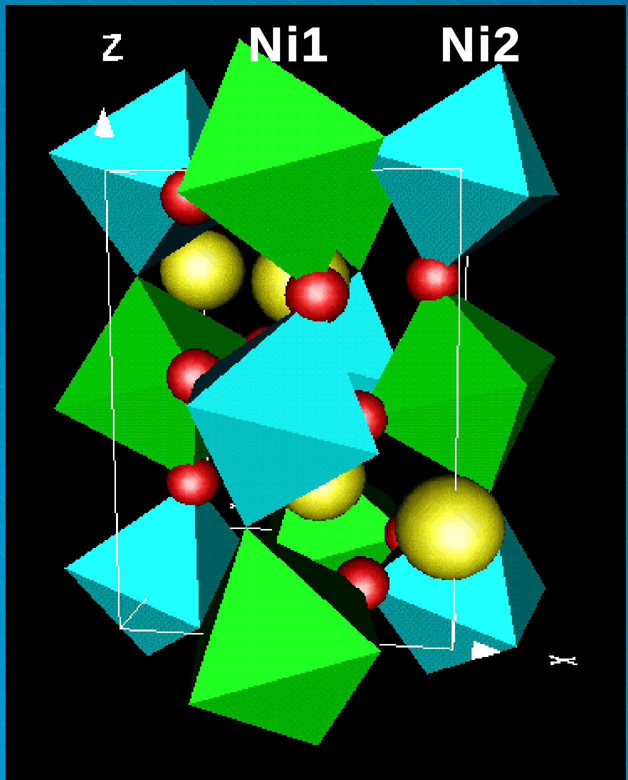
Charge Transfer in $YNiO_3$

Marie-Theresa Fernandez-Diaz et al.

Alan Hewat

Combined ESRF, D1B and D2B data - Alonso J.A. et al (1999) PRL 82, 3873

Metallic Ortho. YNiO3 -> Insulating Mono. YNiO3 T < 582K   Ni valence 3-**d**, 3+ **d**



V(Ni1) = 2.62   V(Ni2) = 3.17

- Double evidence for charge transfer
    - Magnetic superstructure and different moments on Ni-sites
    - Different Ni-O distances around Ni1 and Ni2 sites mean ‘charge transfer’
- Neutrons provide both. But need:
    - High resolution to resolve symmetry
    - High flux to see superstructure

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Neutron Powder Diffraction

Charge Transfer in $YNiO_3$

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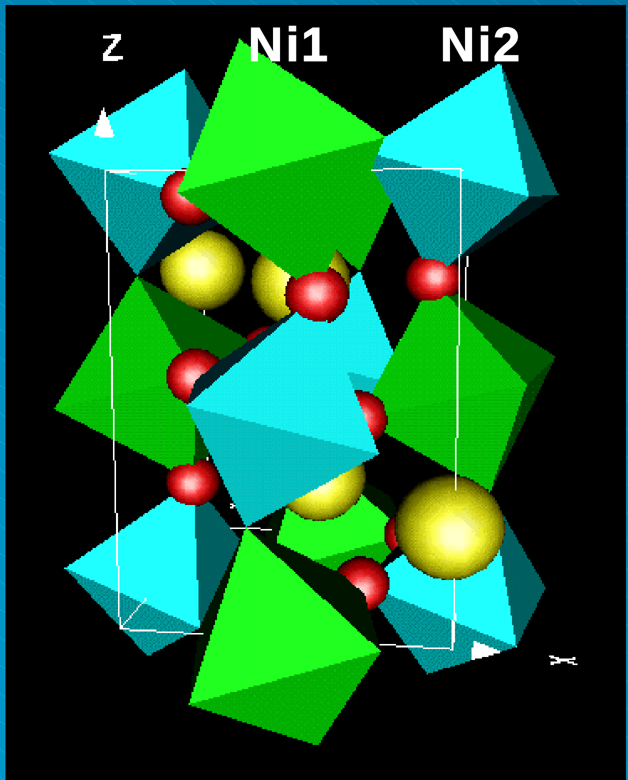
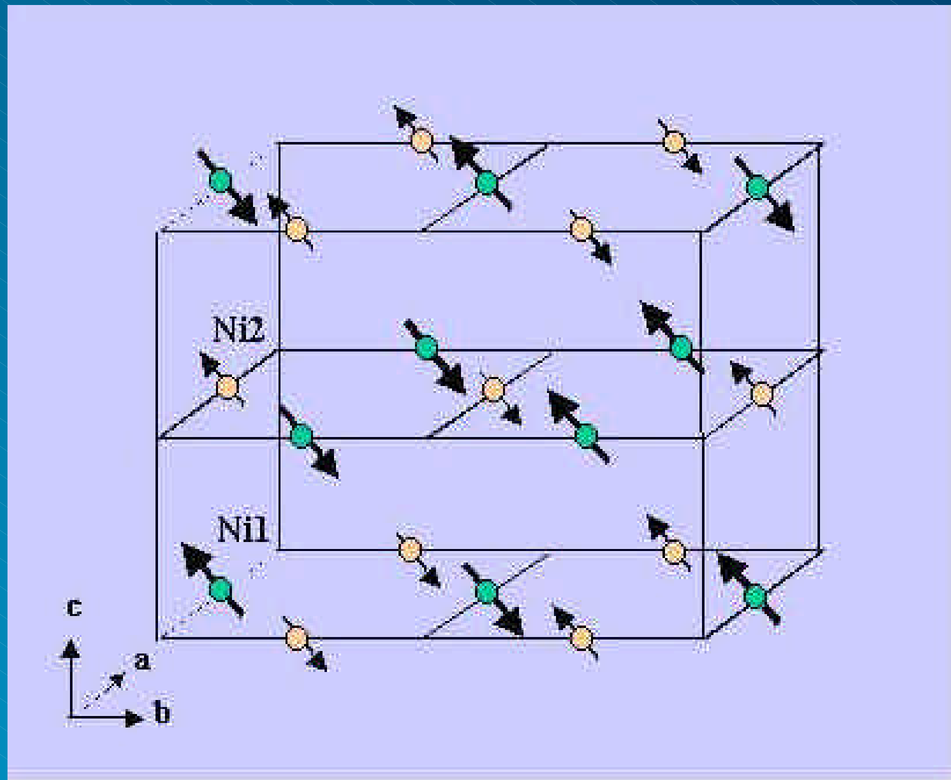
Metallic Ortho. YNiO3 -> Insulating Mono. YNiO3 T < 582K   Ni valence $3-\delta$, $3+\delta$



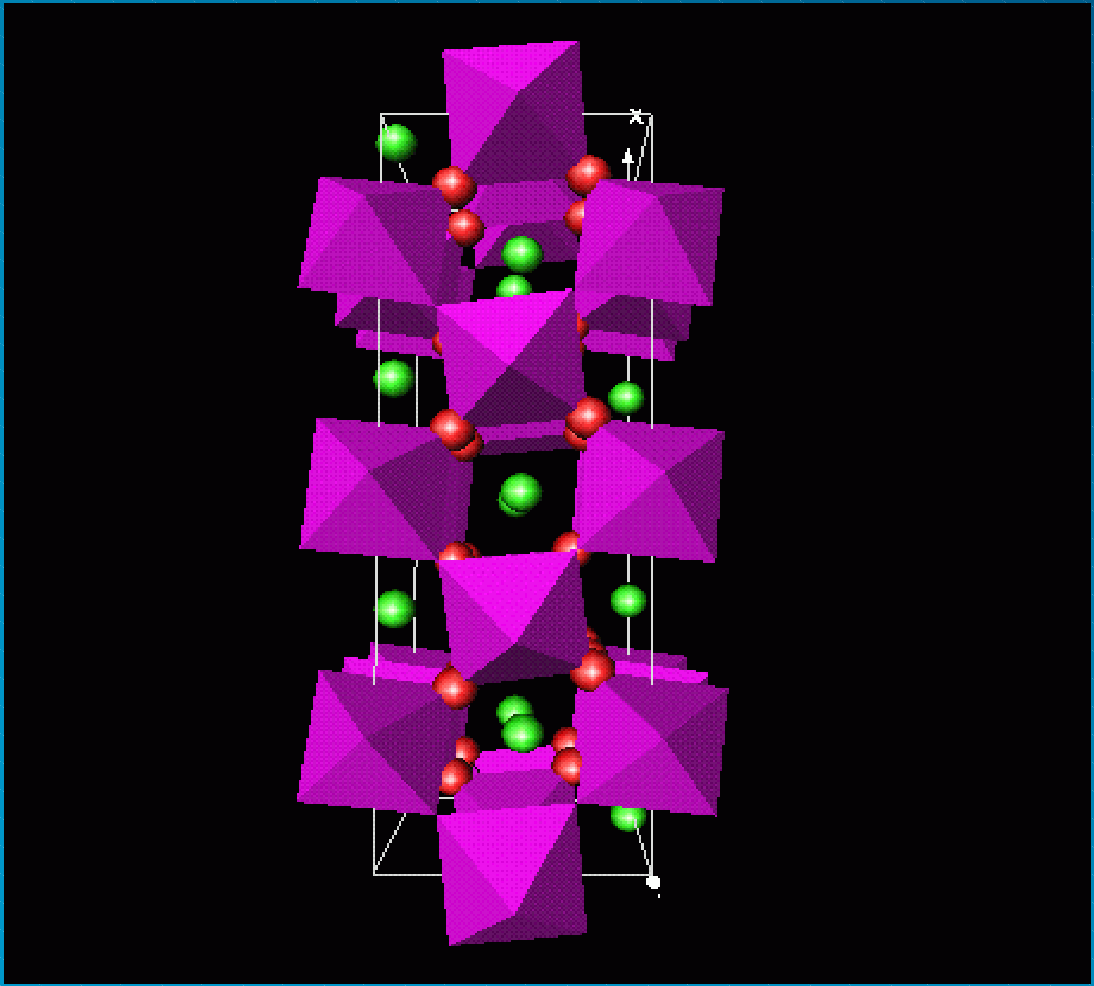


V(Ni1) = 2.62   V(Ni2) = 3.17

M(Ni1) = -1.4 μ_B M(Ni2) = 0.7 μ_B

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

ILL Grenoble
Alan Hewat

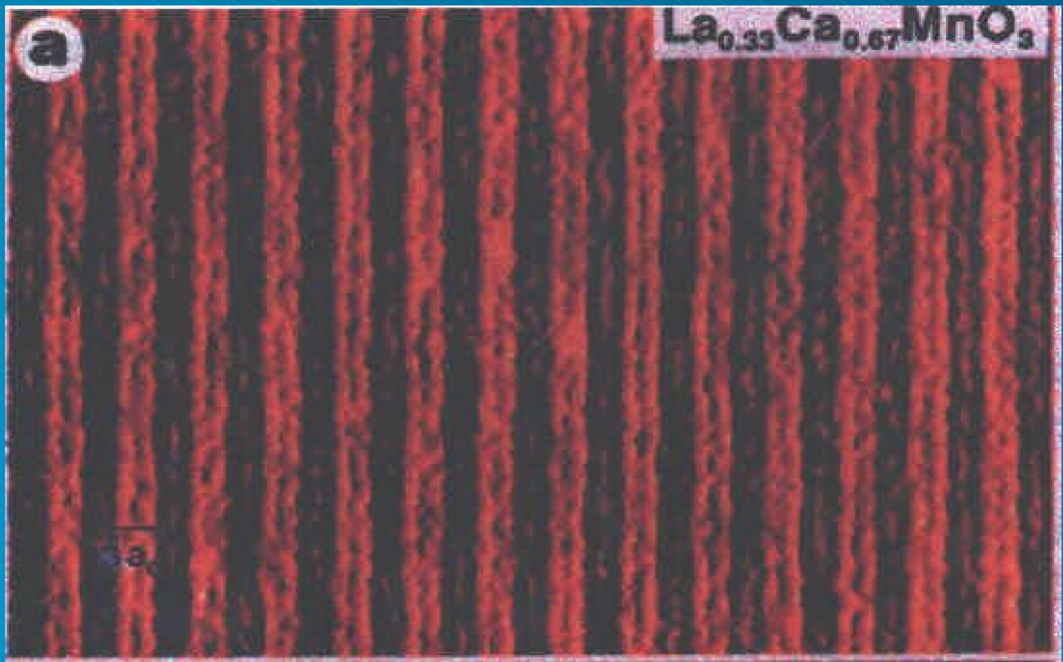


- Very large changes in electrical resistivity with temperature
- cf oxide superconductors
- Mixed valence charge-ordering Mn^{3+}/Mn^{4+}
- GMR effect near room temperature
- Applications to magnetic storage of data (new high density IBM hard disks)

ILL Grenoble
Alan Hewat

GMR Stripes and Charge Ordering

1D-ordering ? Dimensionality important for theory.



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

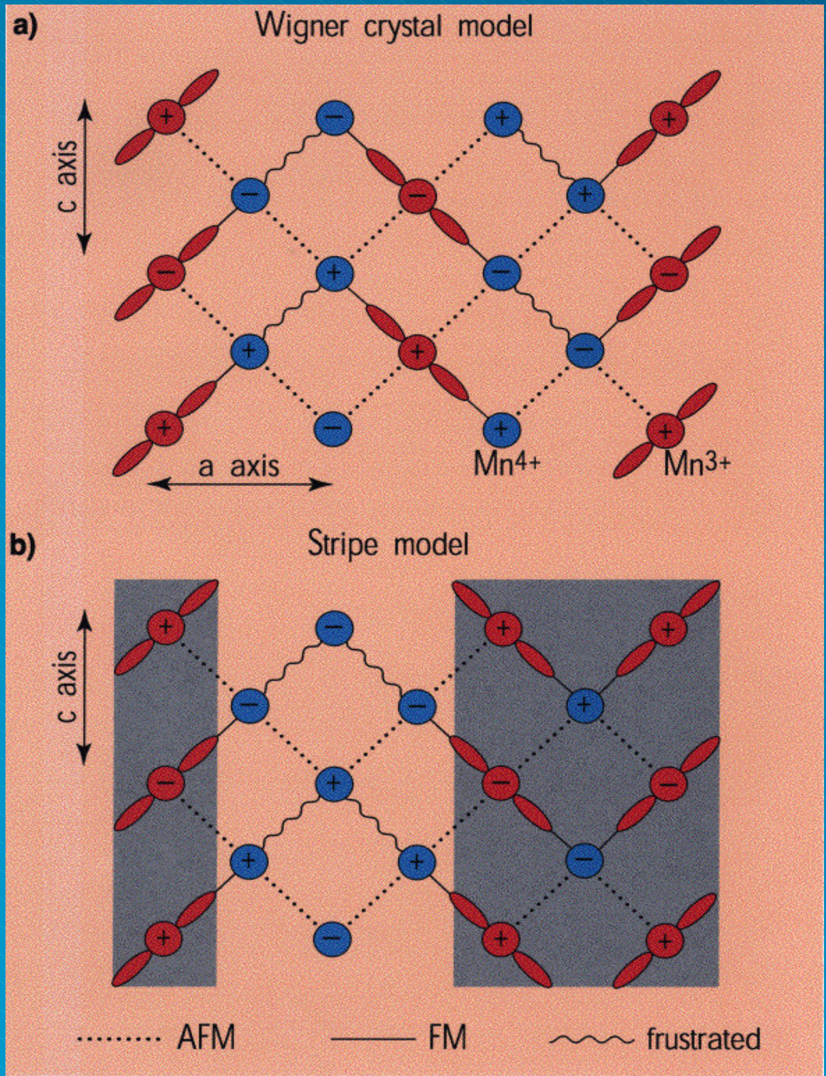
- **Remarkable electron microscope images of 1D stripe pattern in GMR $La_{0.33}Ca_{30.67}MnO_3$**
- **Evidence also for 1D ordering in high-Tc superconductors (Cu^{3+} stripes, spin-ladders etc)**

GMR Stripes and Charge Ordering

ILL Grenoble

Alan Hewat

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

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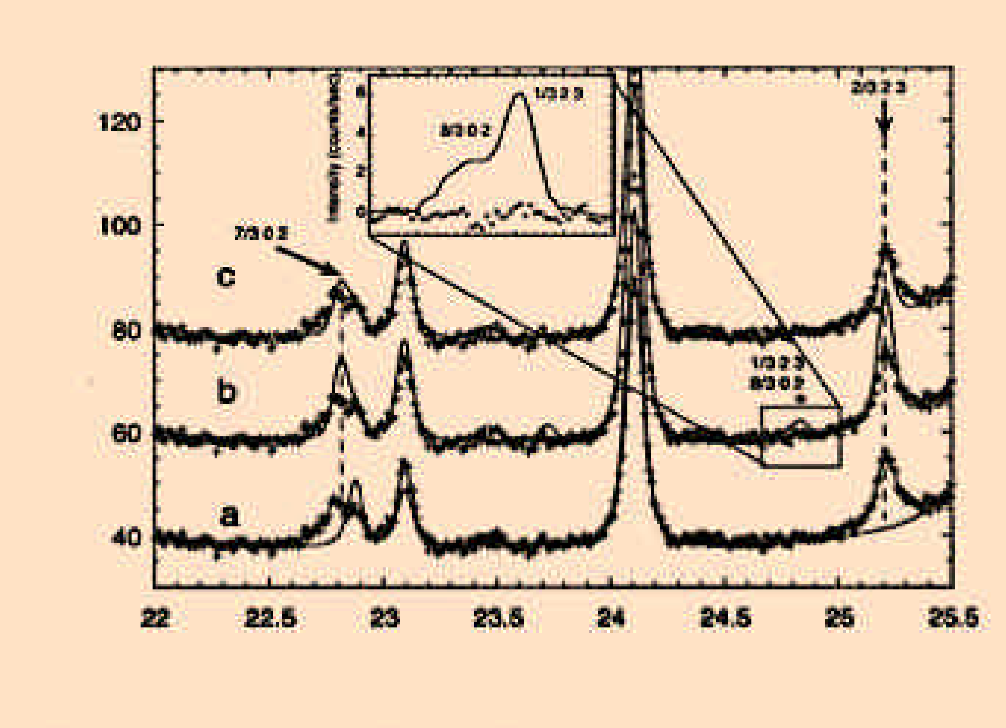
Neutron + Synchrotron Powder Diffraction



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 & ss**
- **High resolution neutron powder data (ILL Grenoble) allows refinement of real structure**
  - a) **Average Structure**
  - b) **Stripe Structure**
  - c) **Wigner Crystal Structure (best fit)**
- **The stripe structure is not supported**

Early Days at ILL Grenoble (1972)

First ILL Powder Diffractometer D1a

Alan Hewat

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

ILL Grenoble

Alan Hewat

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

The Future-Big Detectors super-D2B

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

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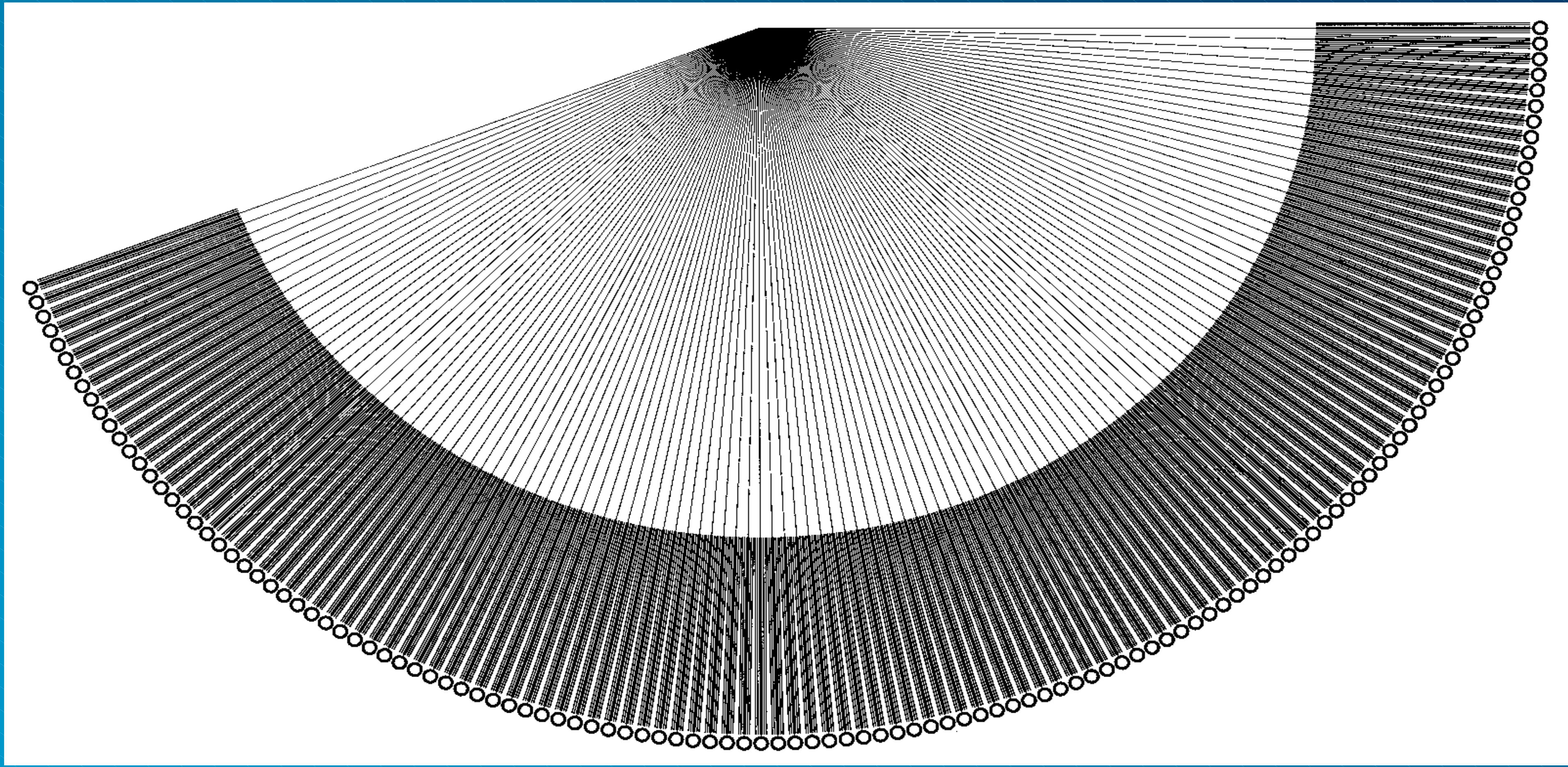

- 2D detector allows both high efficiency & high resolution

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The Future-Big Detectors super-D2B

- 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)**

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The Millennium Programme at ILL
-> Ever Bigger Detectors

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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

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