Neutron Laue Diffraction: A Spotted History, A Scintillating Future

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The high efficiency of modern neutron Laue diffractometers with large area detectors have opened neutron single-crystal diffraction to a wide range of applications in biology, chemistry, materials science, and physics via crystallographic techniques. A survey of pioneering studies of neutron Laue diffraction since the early 1900s at reactors and spallation sources guides us to what we can expect to achieve with upcoming instrumental and data-analysis advances.

In the beginning …

In 1912, following the hypothesis of Max von Laue, Friedrich and Knipping produced the first X-ray diffraction patterns by what would later be called the Laue method: irradiating a stationary crystal by a broad spectrum (X-ray) beam.

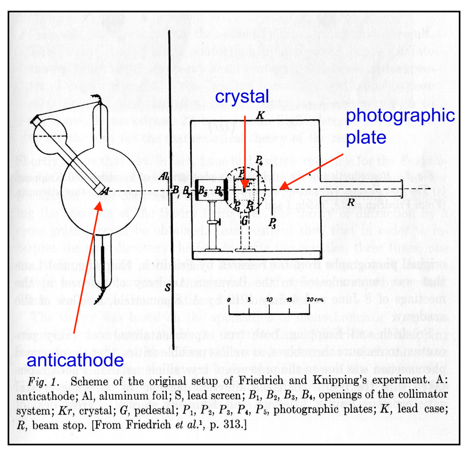
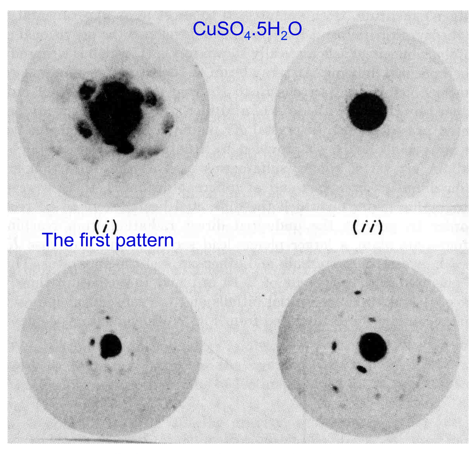


Fig. 1. Scheme of the original setup of Friedrich and Knipping's experiment. A: anticathode; Al, aluminum foil; S, lead screen; B_1, B_2, B_3, B_4, openings of the collimator system; *Kr*, crystal; *G*, pedestal; P_1, P_2, P_3, P_4, P_5, photographic plates; *K*, lead case; *R*, beam stop. [From Friedrich *et al.*[1], p. 313.]

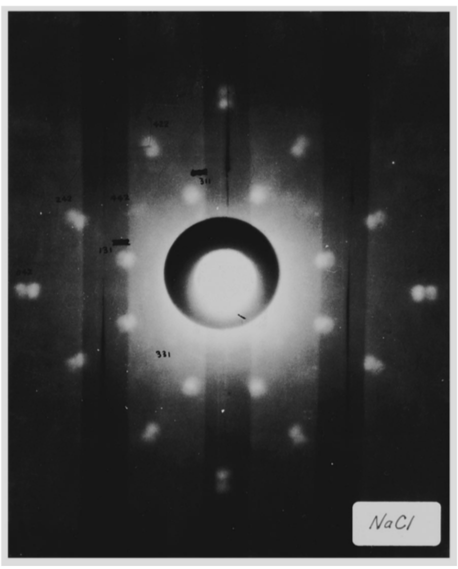


Subsequent improved patterns

Then for nearly 70 years, the method was then used mainly just for crystal alignment and checking quality

In the interim, neutrons join the party

While monochromatic diffractometers and spectrometers using single detectors dominated the first neutron-scattering experiments at Oak Ridge National Laboratory in the late 1940s by **Clifford Schull, Ernest Wollan** and coworkers, there was some interest in exploiting photographic techniques that were so successful for X-rays, e.g in Weissenberg and precession cameras.



Neutron Laue pattern from slightly twinned NaCl: 10 hr exposure

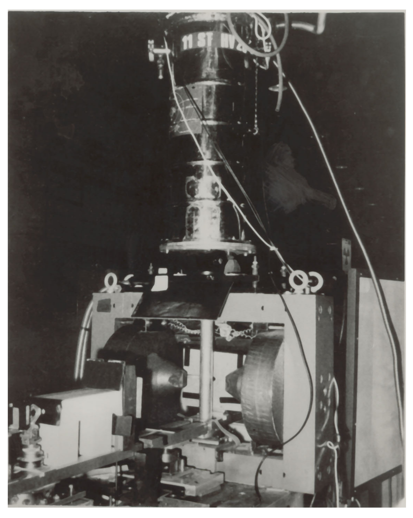
Indium sheets, which emit beta particles upon neutron irradiation, served as a converter to record the **first neutron Laue pattern** (from NaCl) on photographic film.

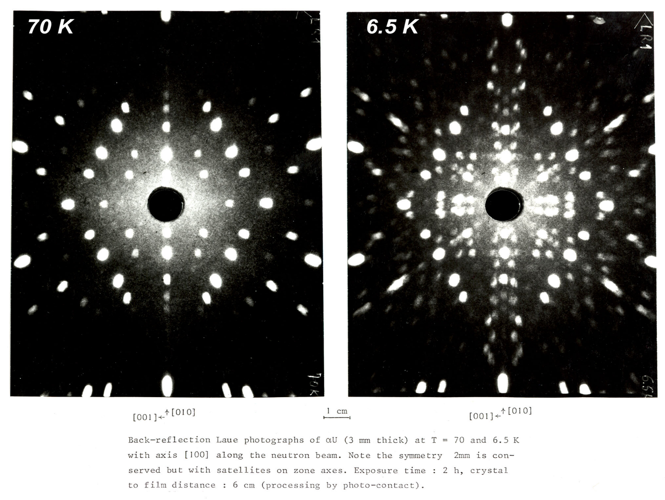
This was also the **first neutron radiograph**, imaging the Scotch Tape strips joining the indium sheets.

One notable exception – Jean-Claude Marmeggi

Marmeggi developed and applied neutron Laue diffraction to challenging physical problems on S42/H24 at the Institut Laue-Langevin (ILL) from 1975 to 2002 using quite limited resources.

Detection method: scintillator and photographic film





Back-reflection Laue photographs of αU (3 mm thick) at T = 70 and 6.5 K with axis [100] along the neutron beam. Note the symmetry 2mm is conserved but with satellites on zone axes. Exposure time : 2 h, crystal to film distance : 6 cm (processing by photo-contact).

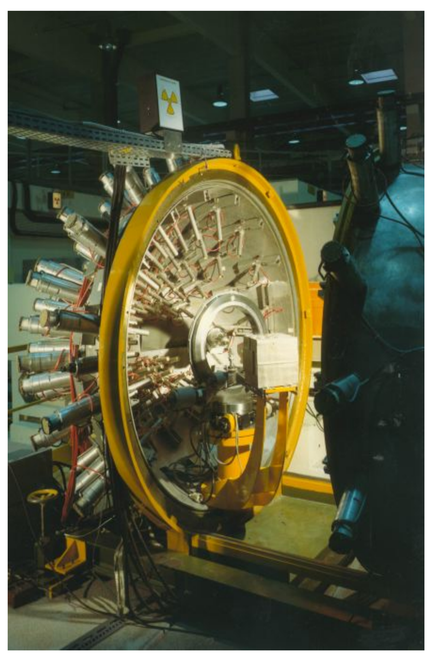
Incommensurate crystal structure of α-U at low temperature

Development of large area detectors

D6 (ILL) – the 'Hedgehog' modified Laue diffractometer

100 single detectors each of which could be positioned individual within a small solid angle.

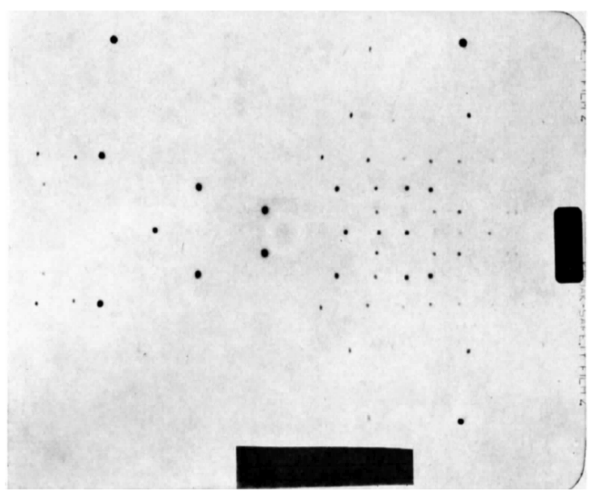
Discontinued in 1975 due to mechanical inaccuracies and limited computing power.



D12 (ILL) – film camera and the modified Laue diffractometer

Modified Laue technique using a bent monochromator to give a dλ/λ ~ 5% with a strong relation between angle and λ.

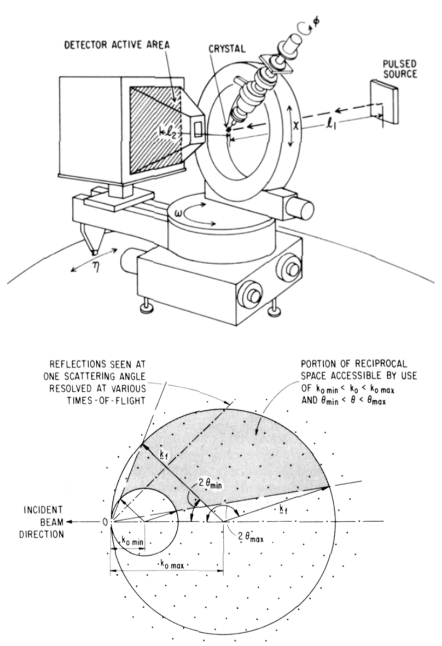
Factor three gain in speed over the world's fastest monochromatic neutron diffractometer in 1977.



Modified Laue photograph from a 0.3 mm^3 histidine HCl H_2O crystal; 30 min exposure; ~350 reflections per pattern

SCD (IPNS) – scintillator-based detector and time-of-flight Laue diffraction

A large solid angle of detection was essential to give a data-collection efficiency comparable to existing monochromatic diffractometers with single detectors at reactor sources (1984).

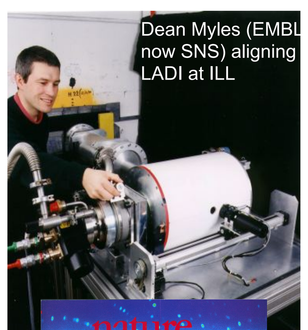


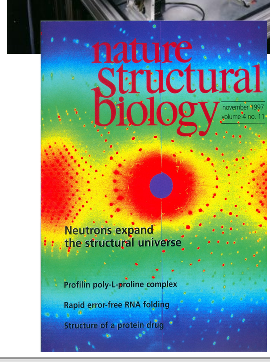
Renaissance at reactor sources near the turn of the century

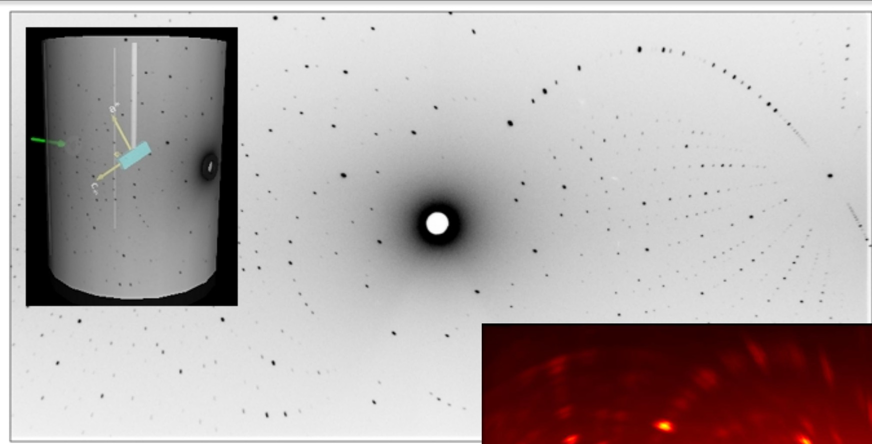
Laue techniques speeded up early protein crystallography at synchrotrons, especially after it was noted that ~82% of the excited spots are single.

The development of neutron-sensitive image plates allowed transfer of the technique to neutron diffraction at reactor sources.

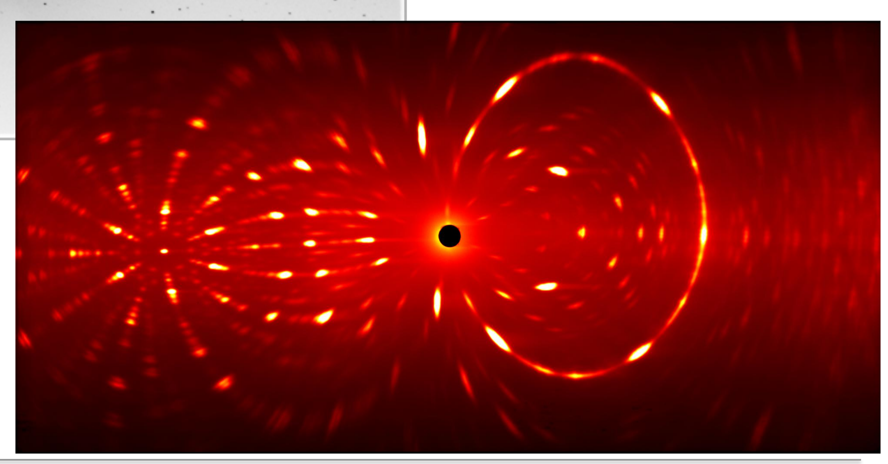
LADI (now LADI-III) and VIVALDI (now DALI) at ILL, KOALA at ANSTO, and IMAGINE at ORNL, have led to a renaissance of the classical Laue technique to apply the unique advantages of neutron diffraction to increasingly smaller and more complex crystalline materials.







Typical Laue pattern from a good quality crystal: hambergite, $Be_2BO_3(OH,F)$

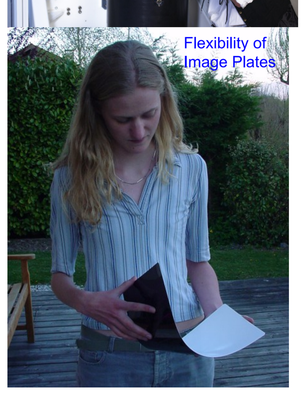


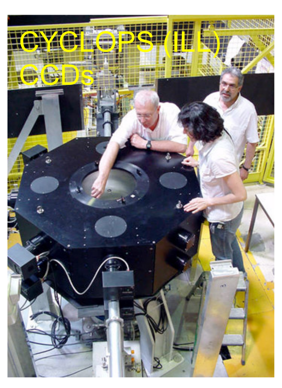
Surveying reciprocal space for new phases: nonadecane-urea

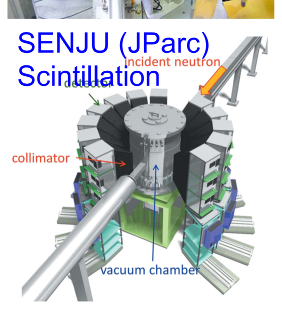
Niche experiments on thermal-neutron Laue diffractometers

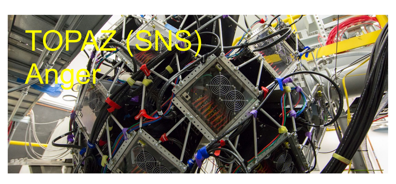
- Conventional crystallography on small single crystals: 0.1 - 1.0 mm^3 sample volume, at 12 - 24 hours per data set is the norm with esds on bond lengths 0.002 - 0.02 Å.
- Rapid data collections on larger crystals with varying temperature or pressure.
- Studies of structural and magnetic phase transitions.
- Volumetric surveys of reciprocal space to search for and characterise incommensurate nuclear and magnetic structures, or to study 1- and 2-D diffuse scattering (both steady state and TOF) and 3-D diffuse scattering (TOF).
- Small crystals in compact pressure cells, e.g. diamond-anvil cells.

Large-area detectors play a key role









Challenges for neutron Laue diffraction

▪ **Detectors**

Long-term commercial source of image plates optimised for neutrons

Long-term commercial source of appropriate CCD-based detectors

▪ **Software**

User-friendly protocols for full and quick analysis of parametric data

Full-pattern fitting

▪ **Training the next generation of users**

Despite the simplicity of the Laue technique, it is not evident to first-time users

▪ **Changing landscape**

14 Laue diffractometers world-wide, but just nine in user programmes

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