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Approaching a Century of Imaging with Neutrons: Significant Advances and Challenges

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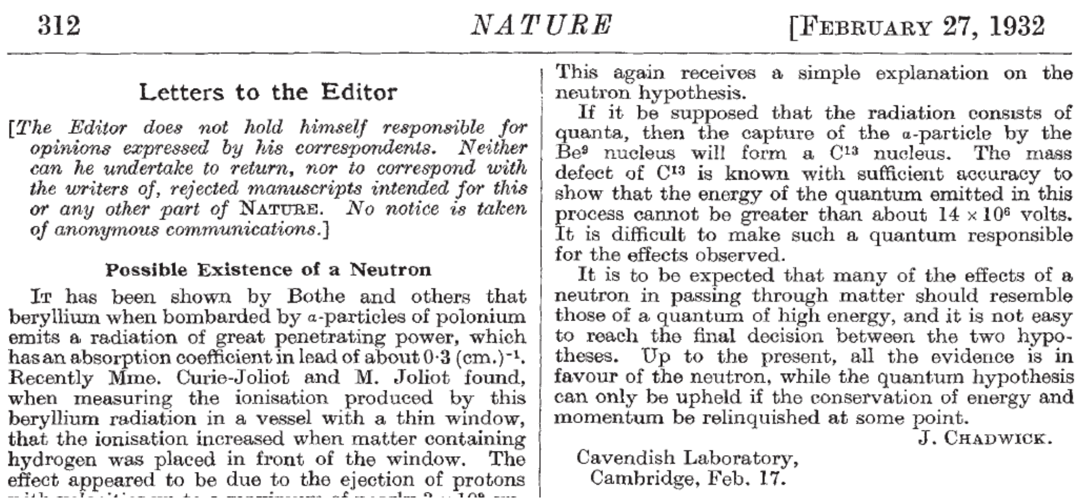
November 5, 2013

Introduction

- I will focus on the events that I feel have had a significant and lasting impact on neutron imaging
  - Many additional individuals have made important contributions to the different areas of neutron imaging
  - The events/advances I discuss here are the ones I feel were the most significant in terms of being the first, were "game changing", and have had a lasting impact
- I apologize in advance if I did not select your contributions for mention!
- I will go through in more or less a chronological order
  - But…I will sometimes comment on later work to add context to the event or advancement being discussed

The Early Years



312 NATURE [FEBRUARY 27, 1932

Letters to the Editor

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Possible Existence of a Neutron

IT has been shown by Bothe and others that beryllium when bombarded by α-particles of polonium emits a radiation of great penetrating power, which has an absorption coefficient in lead of about 0·3 (cm.)$^{-1}$. Recently Mme. Curie-Joliot and M. Joliot found, when measuring the ionisation produced by this beryllium radiation in a vessel with a thin window, that the ionisation increased when matter containing hydrogen was placed in front of the window. The effect appeared to be due to the ejection of protons

This again receives a simple explanation on the neutron hypothesis.

If it be supposed that the radiation consists of quanta, then the capture of the α-particle by the Be^9 nucleus will form a C^{13} nucleus. The mass defect of C^{13} is known with sufficient accuracy to show that the energy of the quantum emitted in this process cannot be greater than about 14×10^6 volts. It is difficult to make such a quantum responsible for the effects observed.

It is to be expected that many of the effects of a neutron in passing through matter should resemble those of a quantum of high energy, and it is not easy to reach the final decision between the two hypotheses. Up to the present, all the evidence is in favour of the neutron, while the quantum hypothesis can only be upheld if the conservation of energy and momentum be relinquished at some point.

J. CHADWICK.

Cavendish Laboratory,
Cambridge, Feb. 17.

- In 1935 Hartmut Kallmann and Ernst Kuhn began to make the first neutron radiographic (NR) images
- Used neutrons from small Ra-Be sources and a small (d,n) neutron generator

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The Early Years

- Carl-Otto Fischer provided an excellent review of the neutron imaging work conducted in Berlin, Germany from 1935-1944
- Kallmann and Kuhn received a joint US patent in January 1940
  - "Photographic Detection of Slowly Moving Neutrons"
  - US Patent 2,186,757

An Aside

- A few slides about neutrons
  - Uncharged particles
  - Produced by bombardment of nuclei with high energy particles or by fission or fusion
  - Neutrons are born with high energies and slow down through interactions with matter
  - In general, the slower the neutron the higher its probability of interaction

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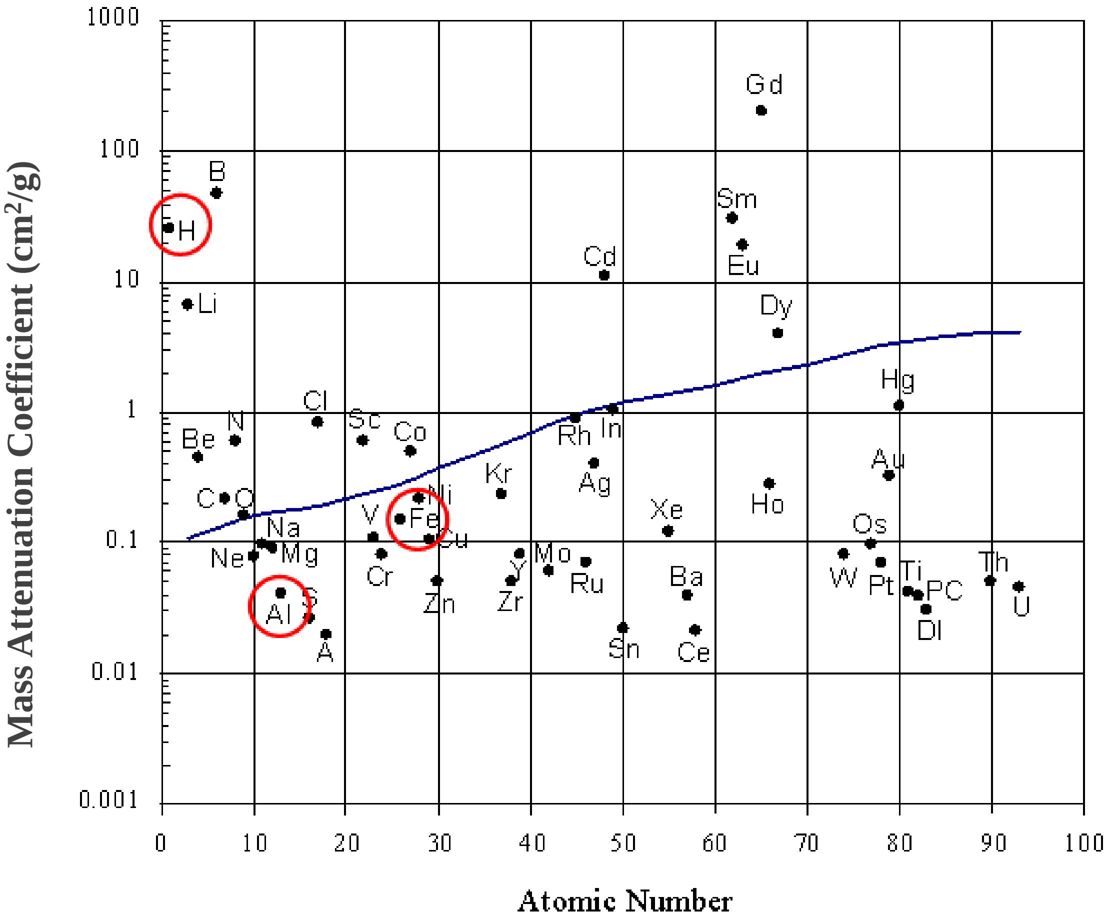
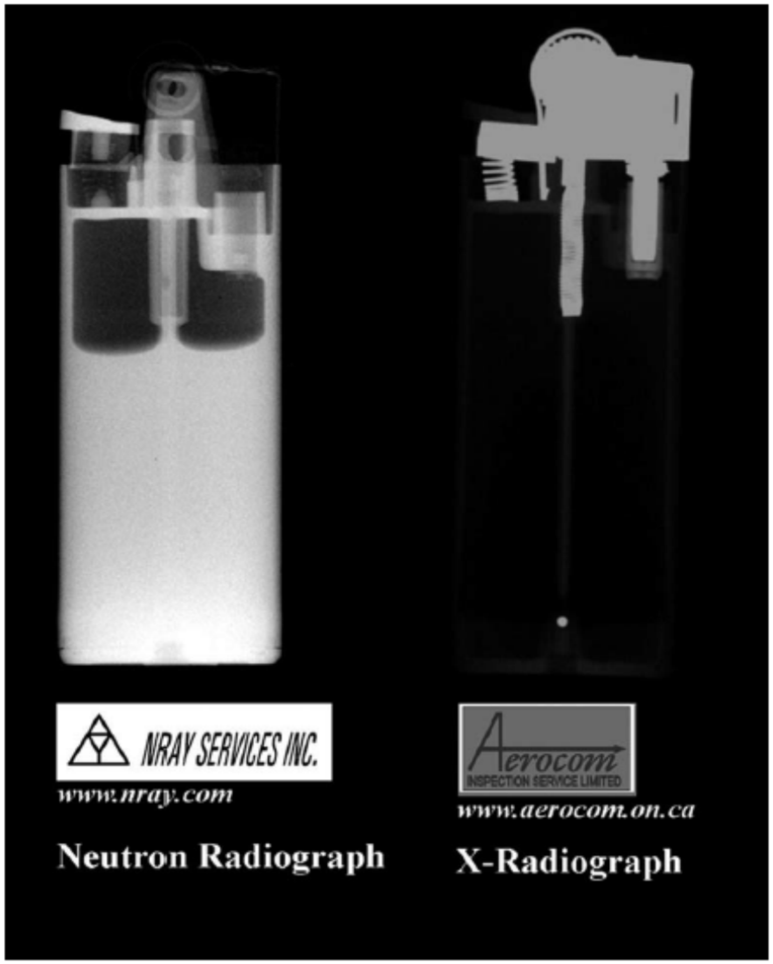
X-ray vs. Thermal (Slow) Neutron Imaging

- Same basic techniques - both are based on absorption and/or scattering of penetrating radiation
- Different interaction mechanisms
  - x rays interact with orbital electrons
  - neutrons interact with nuclei
    - absorption by selected isotopes
    - scattering by low Z isotopes
- Good complimentary NDT techniques

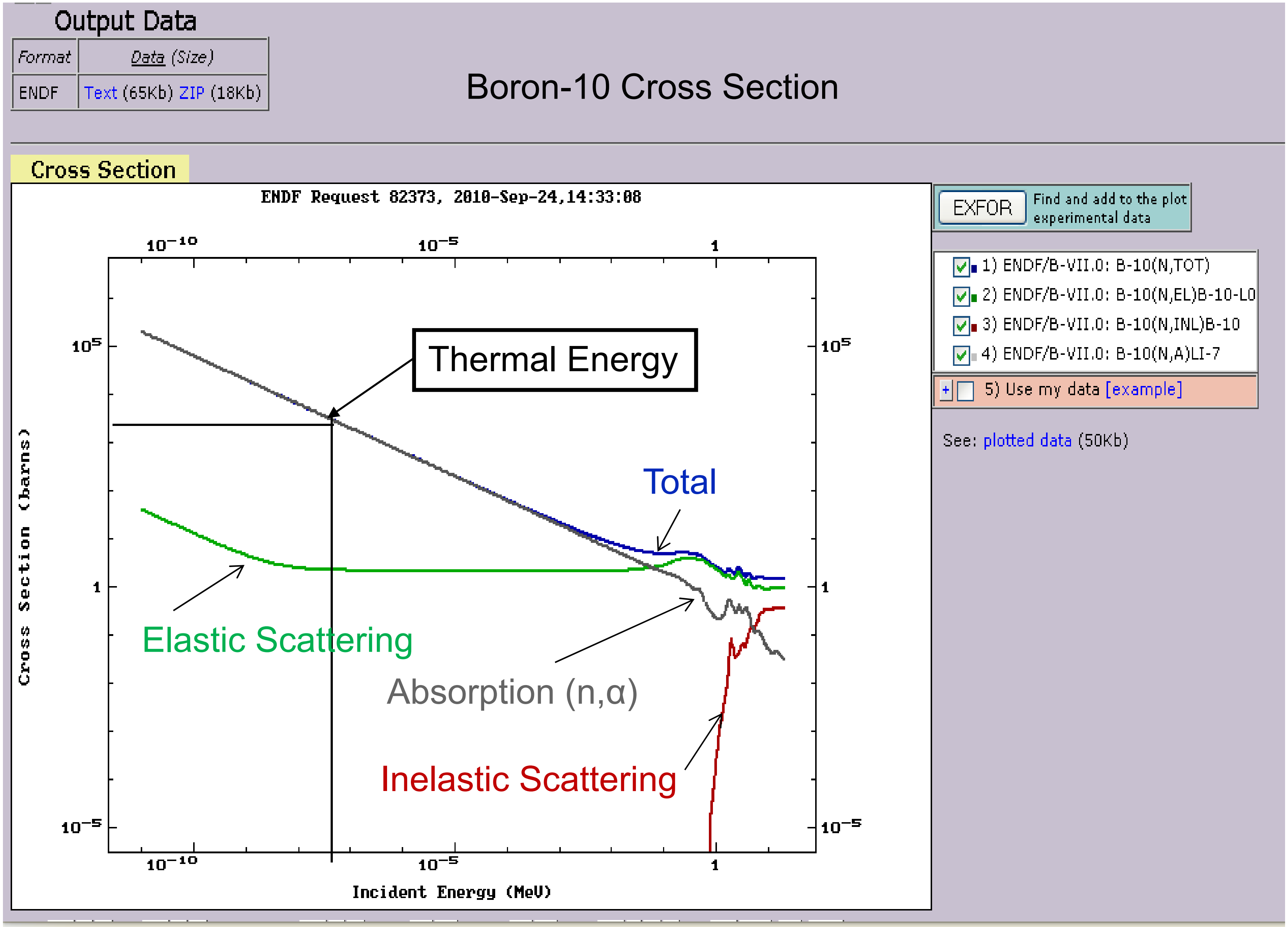
X rays vs. Neutrons



X rays vs. Neutrons



Boron-10 Cross Section



Common Neutron Imaging Technique

- Extract a small solid angle beam of thermal neutrons from the moderator - collimator
- Use a 2D position-sensitive neutron detector to map the neutron intensity pattern behind the test object

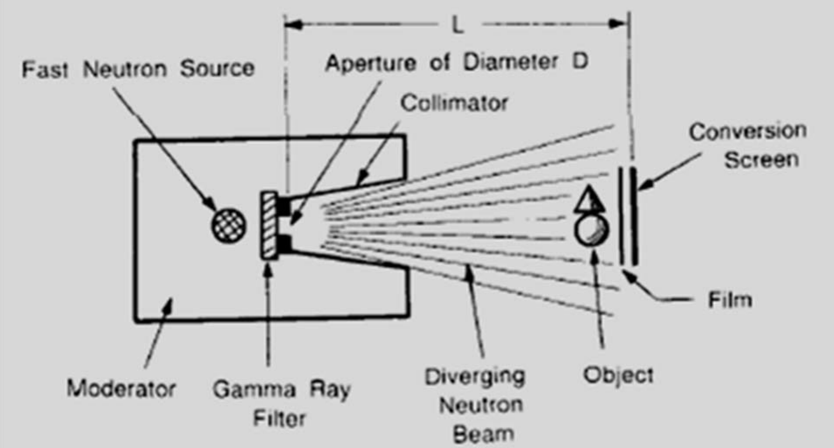
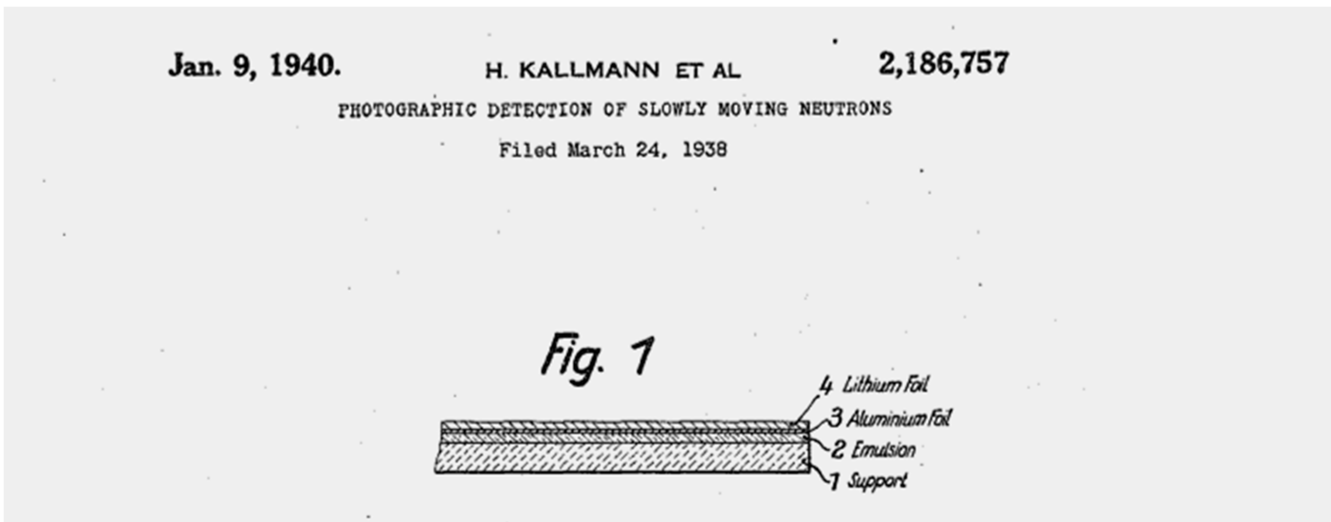


FIG. 1 Typical Neutron Radiography Facility with Divergent Collimator

From ASTM E-748

The Early Years

- Detailed the basic radiographic converter-film system and vacuum cassette


Jan. 9, 1940. H. KALLMANN ET AL 2,186,757

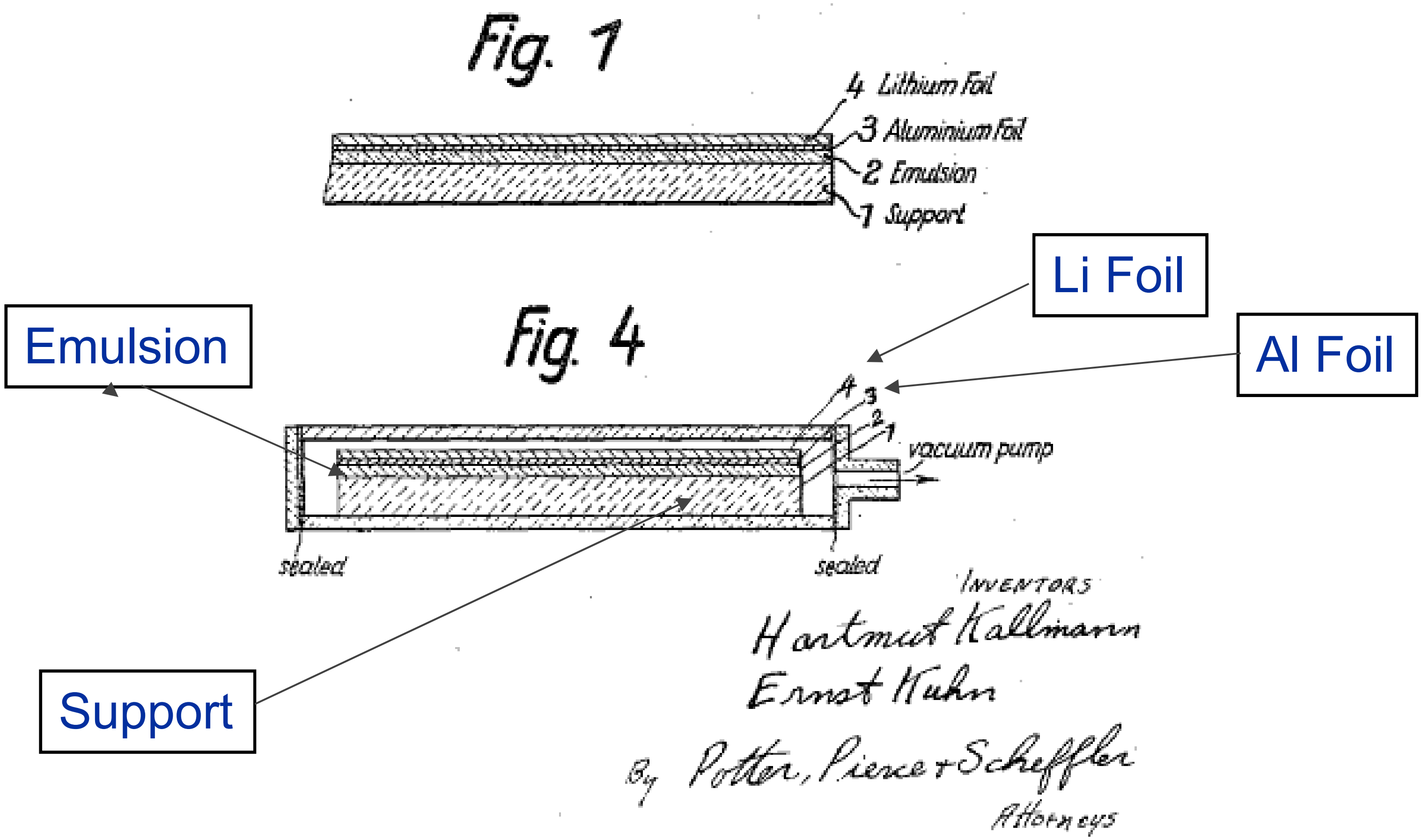
PHOTOGRAPHIC DETECTION OF SLOWLY MOVING NEUTRONS

Filed March 24, 1938

Fig. 1

Kallmann H, Kuhn E. Photographic Detection of Slowly Moving Neutrons. United States Patent 2,186,757, January 9, 1940.

The Early Years



Kallmann H, Kuhn E. Photographic Detection of Slowly Moving Neutrons. United States Patent 2,186,757, January 9, 1940.

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The Early Years

- With a higher intensity neutron accelerator, O. Peter was able to produce radiographs of different objects
- Work ended just before the Soviet army reached Berlin in 1944
- Peter published his work in 1946
- Kallmann published his work in 1948

The Early Years

- It is interesting to note that the Fuji Photo Film Co. and the Japan Atomic Energy Research Institute referenced Kallmann and Kuhn’s patent in their 1998 US Patent 5852301 entitled “Method for forming neutron images“ for their stimulable phosphor neutron imaging sheets

The Early Years

- Few advancements were made until the mid-1950's when J. Thewlis utilized a neutron beam with a flux of between 10^8 to 10^9 n/cm^2-s from the BEPO (British Experimental Pile '0') reactor at Harwell
  - This marked the beginning of the utilization of neutron imaging in practical applications
- As film method improved NR started to become a recognized NDT method
  - NR changed from qualitative to quantitative method

The Early Years

- Opened opportunities to applications in industry and in examination of nuclear fuels

NDE 2011, December 8-10, 2011 15

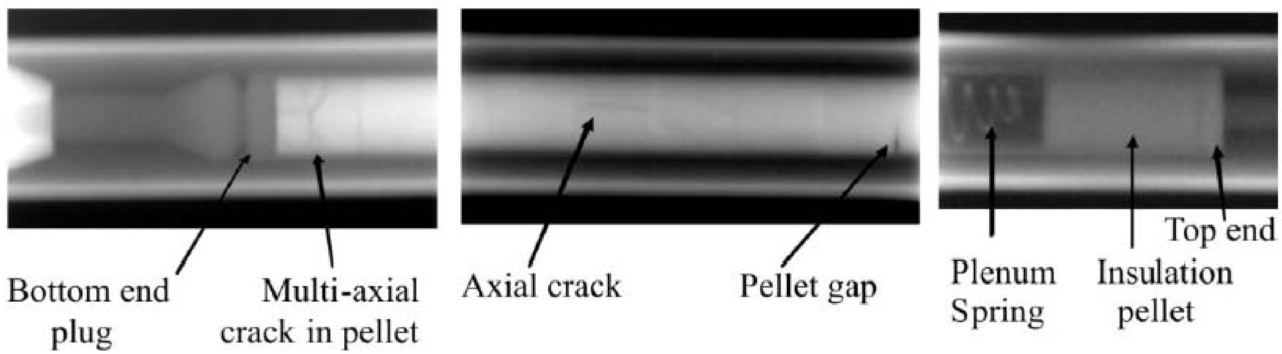

Fig. 7 : The NR shows the ThO_2+4%PuO_2 pellets with multi-axial crack on the left, axial crack and pellet gap in the middle, plenum spring and insulation pellet on the right.

Proceedings of the National Seminar & Exhibition
on Non-Destructive Evaluation
NDE 2011, December 8-10, 2011

NON DESTRUCTIVE EVALUATION OF IRRADIATED NUCLEAR FUEL PINS AT CIRUS RESEARCH REACTOR BY NEUTRON RADIOGRAPHY

J. L. Singh[1], N. K. Mondal[1], M. P. Dhotre[1], K. M. Pandit[1], Anil Bhandekar[1], N. Kumawat[1], Rakesh Ranjan[2], A. K. Sahu[2], N. Ramesh[2] and S. Anantharaman[1]

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The Early Years

- By 1964 there were only two neutron radiography programs in the USA and two in Europe
- One of the first neutron radiography programs in the USA was at the Argonne National Laboratory that was started under the direction of McGonnagle (Berger 1965)
  - Produced what Berger referred to as “one of the earliest reactor quality neutron radiographs”
  - The first application neutron radiographs of radioactive fuel were taken in 1963

The Early Years

- In his 1965 book, Berger stated that the parallel collimator was preferred to obtain an adequate neutron flux

- Barton (1966), working in Grenoble, France, was developing a divergent collimator that would allow large neutron radiographs to be made
- In a subsequent 1967 paper, Barton suggested that the divergent collimator was advantageous for neutron radiography

The Early Years

- Until 1965 much of the neutron radiography focused on using attenuation of thermal neutrons
- Investigation of using neutrons at other energies was beginning
  - Tochilin (1965) performed experimental work on fast neutron radiography at the University of California 60-inch cyclotron, using two different neutron energies
  - Barton (1965) was also investigating the use of different energy neutrons
    - An epithermal radial beam from the Herald reactor at the Atomic Weapons Research Establishment, Aldermaston, England
    - Cold neutrons to examine steel at the same facility

The Early Years

- A number of journal articles and several books were published to describe the neutron imaging methodologies
- Two excellent examples are Berger and Hawkesworth and Walker
  - Berger H. *Neutron Radiography: Methods, Capabilities, and Applications*. Amsterdam - New York: Elsevier Pub. Co.; 1965
  - Hawkesworth MR, Walker J. Review: Radiography with Neutrons. *Journal of Materials Science* 1969;**4**:817-835

The Early Years

- From the mid-1960’s through the 1970’s, a number of nuclear technology research centers became involved in neutron imaging
- Much of the work involved the examination of new and used nuclear fuel
  - The indirect method and the track etch method techniques (Alter patent 1969) were used to examine the highly radioactive fuel
  - The use of these methods has essentially disappeared over the last two decades

The Early Years

- Starting about 1968, a number of facilities began to offer neutron radiography commercial services, a practice that continues today
- The first two reactors to offer such services
  - The Nuclear Test Reactor at the General-Electric Vallecitos Center and
  - The TRIGA type reactor at the Aerotest Company
- A number of reactors at European national laboratories, including the Fontenay-Aux-Roses in France and the Harwell Nondestructive Testing Center in England, also began providing neutron radiography as a service

A Maturing NDT Method

- As neutron imaging moved into the 1970’s, the need to shift from a qualitative or subjective examination method to a quantifiable and standardized method was becoming obvious
- Purchasers of the newly offered neutron radiographic services wanted to have quality control monitors and quantifiable measures in place to be able to verify
  - The reliability of the method to detect defects and
  - Object characteristics for mission critical parts
- Especially true for the USA Apollo space program, where stringent standard methods and recommend practices were already in place for other NDT methods

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A Maturing NDT Method

- Additionally, many practitioners of neutron radiography recognized the relationship between standards and the acceptance of a new NDT method
- Efforts to establish communication among the international community of neutron radiographers resulted in the formation of the Association of Neutron Radiographers (ANR) in 1969 under the leadership of Barton
- Whittemore lead an effort to develop a personnel qualification standard under the authority of the American Society for Nondestructive Testing (ASNT), SNT-TC-1A that was first published in 1974

A Maturing NDT Method

- Concurrently, work began in the then American Society for Testing and Materials (ASTM) to develop image quality indicators, terminology, and recommended practices
- ASTM Committee E-7 on Nondestructive Testing established a Section E07.01.02 on Neutron Radiography under the chairmanship of E. L. Criscuolo
- Out of this Section came two indictors that became internationally recognized: the Beam Purity Indicator (BPI) and the Sensitivity Indicator (SI)

A Maturing NDT Method

  - The Beam Purity Indicator (BPI) and the Sensitivity Indicator (SI) devices received ASTM approval in 1975 and are still in use today
- Development and maintence of neutron imaging standards continues today in ASTM International's Subcommittee E07.05 Neutron Radiology
- Efforts were made to establish neutron imaging standards in the International Organization for Standardization (ISO) under committee ISO/TC/135/SC 5/WG 4 in the 1990's, but the effort did not result in any ISO standards

A Maturing NDT Method

- Neutron imaging was growing rapidly internationally, with facilities in the United States, in many European countries and in Japan
- In 1973, the first conference relating specifically to neutron imaging, "Radiography With Neutrons," was held at the University of Birmingham (Hawkesworth)
  - The proceedings of this conference were published in 1975 by Hawkesworth
  - This conference played an important role in bringing the neutron imaging researchers and practitioners together as a community

A Maturing NDT Method

- Dynamic imaging was being performed by those working in x-ray radiography and interest quickly spread to the neutron radiography community
- In 1966, Berger reported the results of his tests of what was then referred to as a neutron television system.
  - The system consisted of a neutron sensitive image intensifier tube coupled to a vidicon television camera with 525 lines and a 30 frame per second frame rate
  - The system was capable of 0.5 mm resolution

A Maturing NDT Method

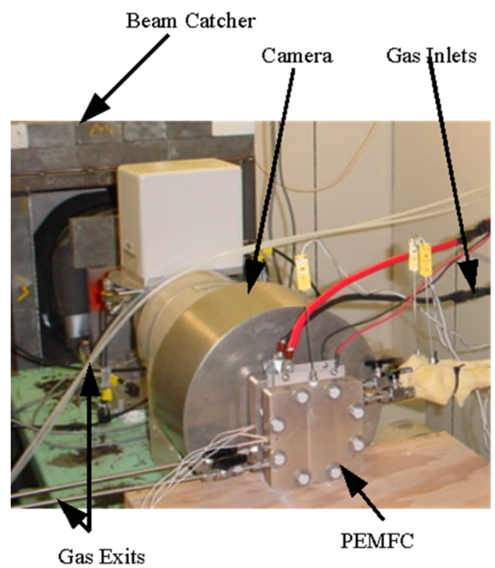
- In Japan, S. Kawasaki (1968) was working with a collimated thermal beam of 10^4 n/cm^2-s from a high yield (d,t) neutron generator
  - The system utilized a multi-stage image intensifier
  - Spatial resolution was approximately 1 mm with a neutron flux of 5 X 10^3 n/cm^2-s
- In the United Kingdom, Hendry (1969) and his colleagues utilized the Materials Testing Reactor neutron radiography unit at Dounreay with an image intensifier to provide “immediate radiography” by direct viewing of a scintillating plate

A Maturing NDT Method

- High speed neutron radiography was accomplished by Bossi, Robinson and Barton (1983) at the Oregon State University's TRIGA reactor
  - Utilized a reactor pulse and a high speed camera
    - The high-speed motion neutron radiography system incorporated a pulsing reactor, a low L/D ratio collimator, a scintillator screen, an image intensifier, and a high speed film video camera
    - The camera was synchronized to the reactor pulse
    - An exposure time of 40 μs per frame was obtained at a frame rate of 10000 frames per second

A Maturing NDT Method

- By the mid-1970's "real-time" neutron imaging systems were commercially available
  - Most based on an electrostatic image intensifier, a gadolinium oxysulfide scintillator (GOS) and a isocon or newvicon television tube
- Thomson-CSF (1983) produced a robust neutron intensifier tube that was widely used through the 1990's for dynamic neutron imaging



A Maturing NDT Method

- In the mid-1970's, interest began to build to perform computed axial tomography with neutrons
- Early efforts demonstrated the technique's possibilities, but were hindered by low beam intensities, the lack of high-resolution digital sensors, and slow computers with limited memory space
  - Most facilities were using a well collimated beam, a rotating sample holder, and x-ray film as the sensor
- Schlapper and his collegues reported their neutron tomography investigations at the Missouri University Research Reactor in 1977

A Maturing NDT Method

- The preliminary results were poor in comparison to today's neutron computed tomography (NCT) reconstructions
- Rapidly growing computational capabilities coupled with the new real-time imaging systems overcame many of these limitations
- Imaging by Matsumoto and Krata using a neutron generator and a new "neutron television" system reported in 1981 at WCNR1

A Maturing NDT Method

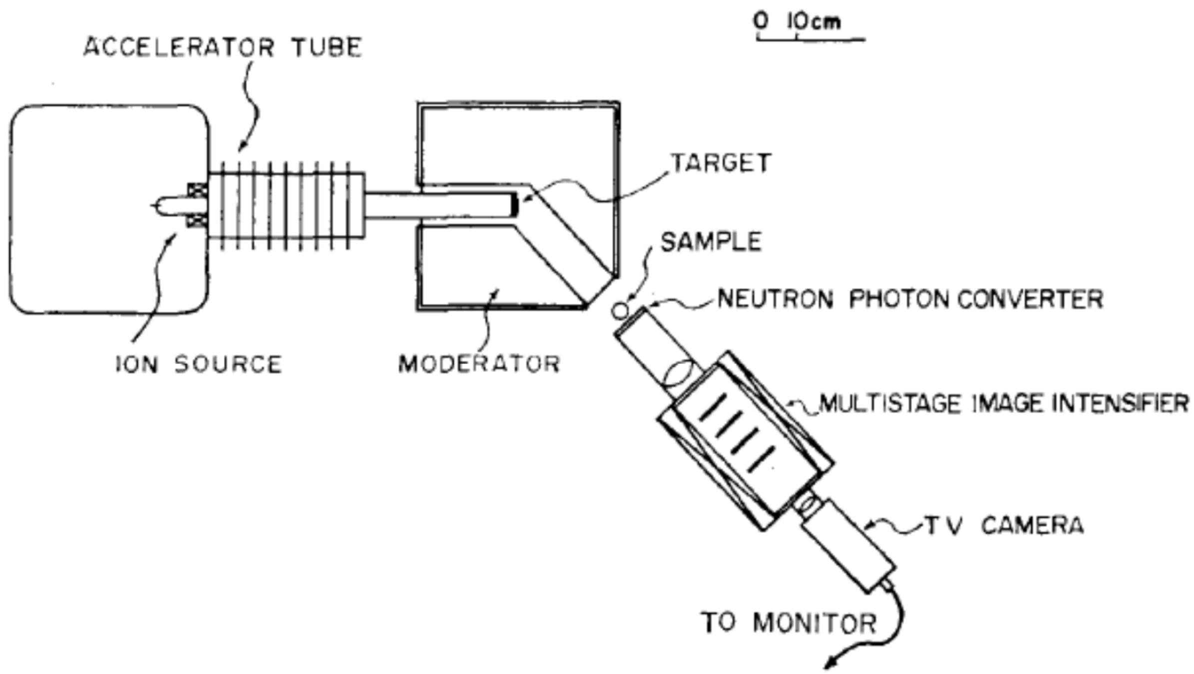


Fig. 1. The schematic drawing of the thermal neutron television system.

Matsumoto G, Krata S, The Neutron Computed Tomography. In: Barton JP, Von Der Hardt P, editors. *Neutron Radiography, Proceedings of the First World Conference*, San Diego, CA: D. Reidel Pub. Co.; 1983, p. 899-906.

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A Maturing NDT Method

- The number of researchers working in NCT grew quickly
  - For example, in the proceedings of the first World Conference on Neutron Radiography (WCNR) in 1981, there was one paper on NCT while at the second WCNR held in 1986, there were ten papers on NCT.
- The NCT technique opened a new field in neutron imaging that is still expanding today

http://www.psi.ch/niag/TomographyEN/igp_300_horsefly.png

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The Mid-1980's to Mid-2000's

- As neutron imaging moved into the 1980's more researchers were joining the neutron imaging community
- An event that was to have a significant and long-term impact on neutron imaging was the establishment of the Neutron Radiography World Conference (WCNR) series and the publication of the conference proceedings by Barton

The Mid-1980's to Mid-2000's

- With the exception of the 1973 University of Birmingham "Radiography with Neutrons" conference and several ASTM symposia
  - Active participants and those interested in learning about the technique had little opportunity to gather to exchange ideas, new techniques and applications
- Papers were usually published in either the journal associated with the application or in an NDT journal not easily accessible outside the NDT community

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The Mid-1980's to Mid-2000's

- MacGillivray and Brenizer (1989) decided that it would be valuable to hold a second smaller meeting that:
  - promoted discussion among the active practitioners of neutron imaging and
  - resulted in journal papers on neutron imaging that would be readily accessible through indexing databases
- The first International Topical Meeting on Neutron Radiograph focused on System Design and Characterization was held in Pembroke, Ontario, Canada (1990)
  - The meeting, while successful, it did not meet its goal of publishing its proceedings in an indexed journal.
- ITMNR2 (1995) in Japan, maintained the same theme, with the proceedings were published in Nuclear Instrumentation and Methods in Physics

The Mid-1980's to Mid-2000's

- The number of practitioners and neutron imaging facilities began to decline from the mid-1980's through the 1990's
  - Activity and participation fell in all neutron imaging areas
- There were two major factors that negatively impacted neutron imaging research and service work
  - New competitive NDT techniques were developed that were portable, less expensive, required a shorter examination time and did not activate the examined materials
  - The international climate that did not fund or support nuclear activities

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The Mid-1980's to Mid-2000's

- Renewed interest in neutron imaging in the mid-1990's was sparked by
  - New facilities
  - New imaging devices
  - Older techniques improved by advances in computer processors, memory storage and imaging sensors
- Five new neutron imaging facilities became operational between 1994 and 2004

The Mid-1980's to Mid-2000's

- There have been three technology achievements that have had a dramatic change in neutron imaging in the last two decades
  - None of which were initially designed to enhance neutron imaging

1) The dramatic improvement in computer processor and bus speeds, coupled with the availability of large, fast active memory and data storage devices:
   - Enabled data analysis, image processing and image reconstructive techniques that were either not possible (or at least impractical) only ten years ago
   - Costs for the fast and powerful computers required for data collection and processing, once a major facility expense, are now quite low compared to the imaging facility hardware

The Mid-1980's to Mid-2000's

2) The on-going development of CCD imaging chips and low-light cameras with multiple millions of pixels
   - CCD cameras have increased the achievable spatial resolution in digital neutron images, especially large format images
   - Not only has pixel size been reduced, but
     - The transfer rate of image data has increased
     - The sensitivity to low-light level is quite high
     - The solid-state cooling has reduced black current noise
     - The cost has dropped steadily over the last decade
   - Easy to set up a very acceptable neutron imaging system quickly and inexpensively

The Mid-1980's to Mid-2000's

3) The development of improved imaging devices for the medical community has had a major impact on neutron imaging
   - Photostimulable phosphor plates provide an alternative for x-ray film
   - Manufactures have called this technique Computed Radiography (or less frequently Computed Radiology)
   - The advantages of the CR systems are clear (Kobayashi, 2011)
     - A linear response over a wide exposure range
     - Plates are reusable
     - No processing chemicals are required
     - No hazardous wastes are produced
     - Plates can be produce a large-format digital image

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The Mid-1980's to Mid-2000's

- CR systems are having a dramatic effect on neutron imaging
  - Service providers
  - The customers that rely on neutron radiography (film) for quality assurance of their parts and products
- Single emulsion high resolution films used in direct neutron radiography have been or will soon be discontinued
- Currently no standards or image quality devices available for use with neutron CR or digital systems

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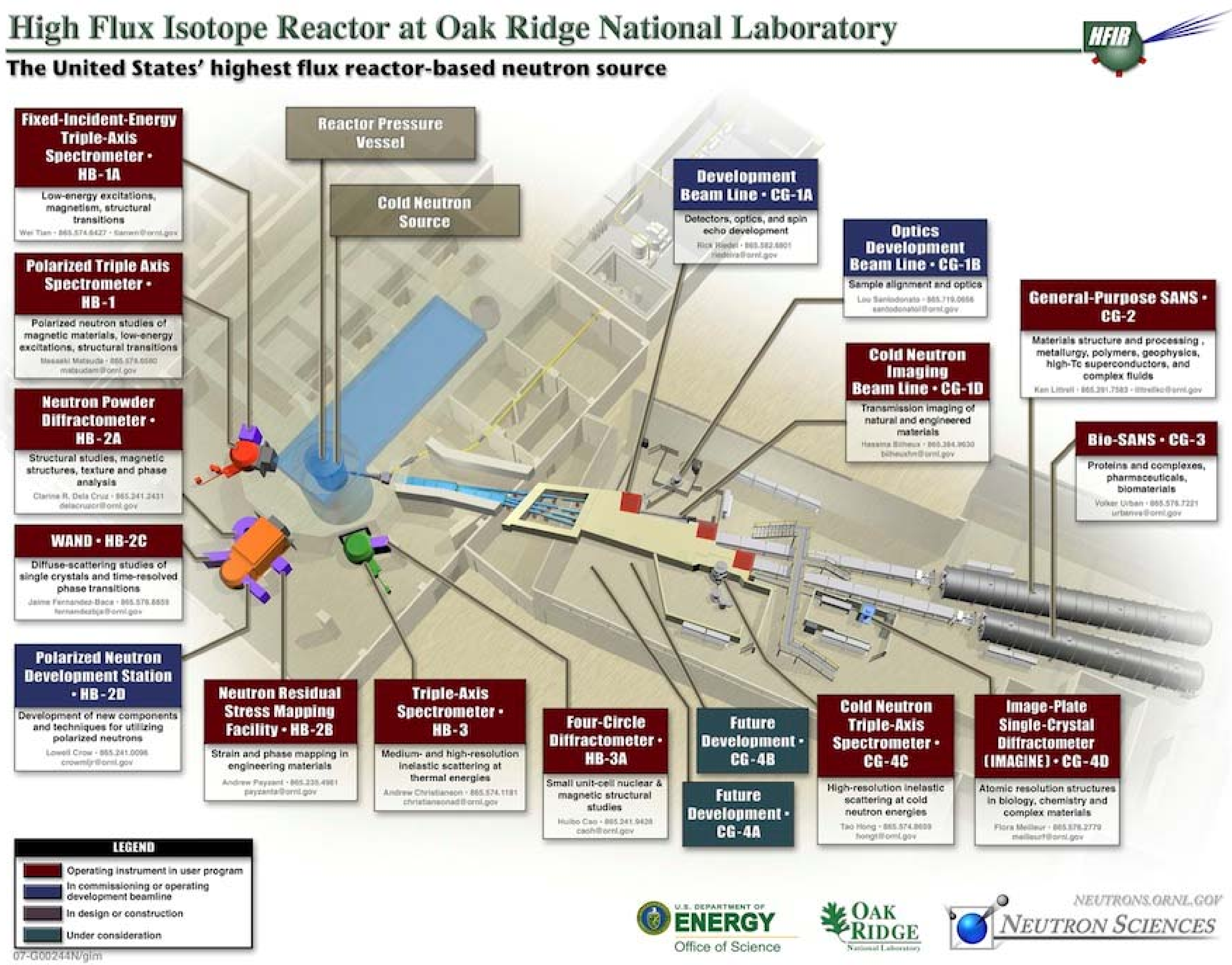
The Mid-2000's to Present

- New approaches, sensors and facilities have been developed
  - Amorphous silicon sensors and MCP's
    - Improved temporal resolution
    - A reasonably large imaging area for dynamic imaging
  - Have not yet had the wide-ranging impact of the three mentioned above

The Mid-2000's to Present

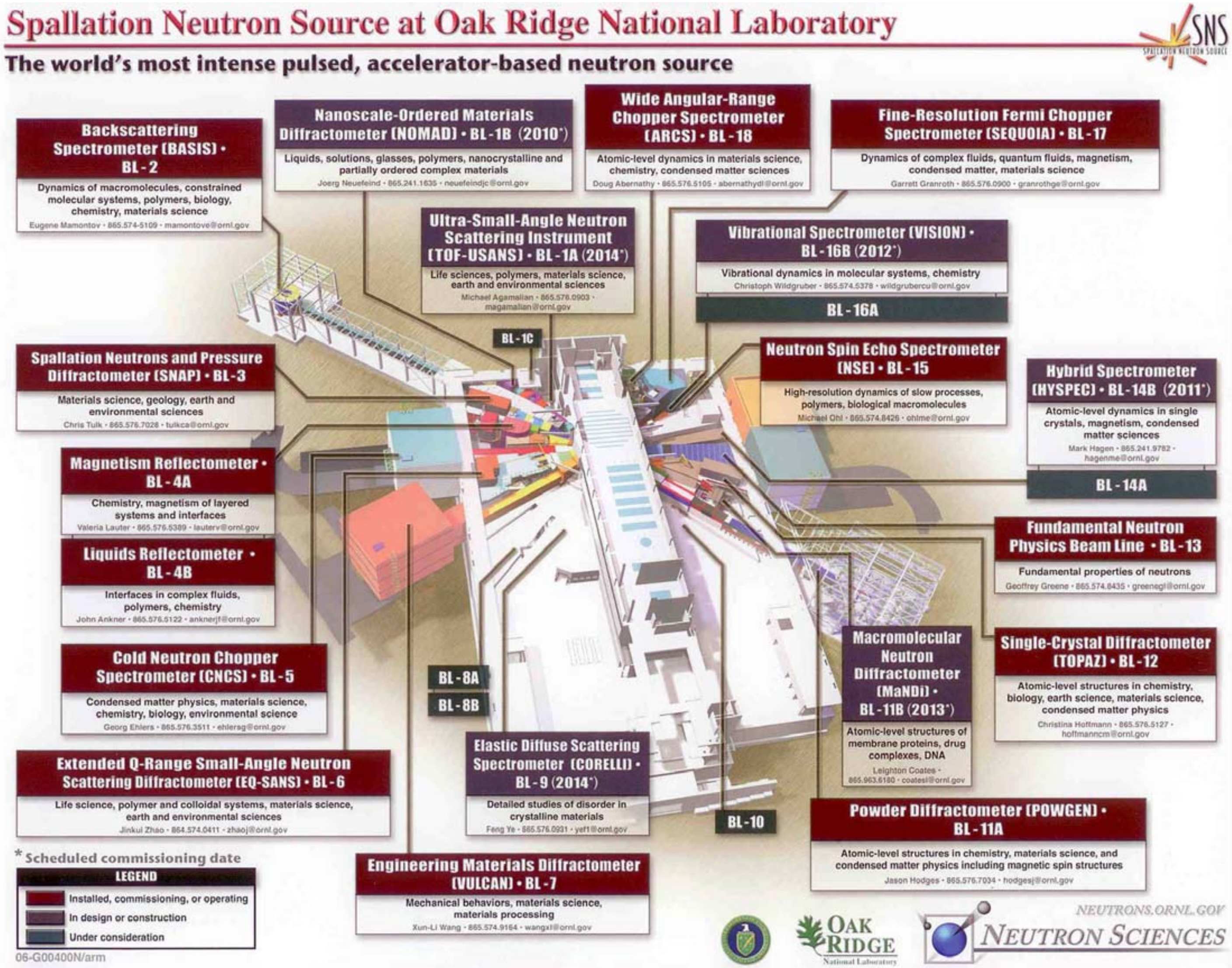
- Neutron imaging is expanding and allowing visualization of neutron scattering information


High Flux Isotope Reactor at Oak Ridge National Laboratory
The United States' highest flux reactor-based neutron source
HFIR
Fixed-Incident-Energy Triple-Axis Spectrometer • HB-1A
Low-energy excitations, magnetism, structural transitions
Polarized Triple Axis Spectrometer • HB-1
Polarized neutron studies of magnetic materials, low-energy excitations, structural transitions
Neutron Powder Diffractometer • HB-2A
Structural studies, magnetic structures, texture and phase analysis
WAND • HB-2C
Diffuse-scattering studies of single crystals and time-resolved phase transitions
Polarized Neutron Development Station • HB-2D
Development of new components and techniques for utilizing polarized neutrons
Reactor Pressure Vessel
Cold Neutron Source
Neutron Residual Stress Mapping Facility • HB-2B
Strain and phase mapping in engineering materials
Triple-Axis Spectrometer • HB-3
Medium- and high-resolution inelastic scattering at thermal energies
Four-Circle Diffractometer • HB-3A
Small unit-cell nuclear & magnetic structural studies
Future Development • CG-4B
Future Development • CG-4A
Cold Neutron Triple-Axis Spectrometer • CG-4C
High-resolution inelastic scattering at cold neutron energies
Image-Plate Single-Crystal Diffractometer (IMAGINE) • CG-4D
Atomic resolution structures in biology, chemistry and complex materials
Development Beam Line • CG-1A
Detectors, optics, and spin echo development
Optics Development Beam Line • CG-1B
Sample alignment and optics
Cold Neutron Imaging Beam Line • CG-1D
Transmission imaging of natural and engineered materials
General-Purpose SANS • CG-2
Materials structure and processing, metallurgy, polymers, geophysics, high-Tc superconductors, and complex fluids
Bio-SANS • CG-3
Proteins and complexes, pharmaceuticals, biomaterials
LEGEND
Operating instrument in user program
In commissioning or operating development beamline
In design or construction
Under consideration
07-G00244N/gim
U.S. DEPARTMENT OF ENERGY
Office of Science
OAK RIDGE
National Laboratory
NEUTRONS.ORNL.GOV
NEUTRON SCIENCES

Neutron Imaging at SNS

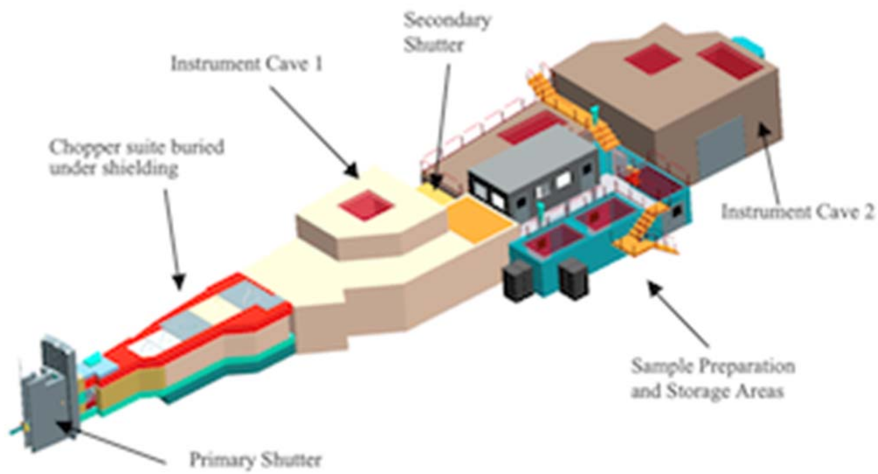
- Oak Ridge National Laboratory – Spallation Neutron Source (SNS)

SNS Instrument Layout



Neutron Imaging at SNS

- Versatile Neutron Imaging Instrument at SNS (VENUS)
- VENUS approved to go on last open beam line in target building – BL10
- Range of cold, epithermal and fast neutrons at SNS will give users of VENUS access to novel imaging methods
- The time-of-flight (TOF) neutrons will offer easy and cost-efficient access to energy-selective imaging
  - Use of neutron scattering Bragg features for improved contrast and identification of phases in an absorption image
- The high peak flux will be used for stroboscopic imaging of repetitive or cyclic motions and is synchronized to a selected neutron energy range for enhanced image contrast



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World Neutron Imaging Facilities

E.H. Lehmann et al. / Nuclear Instruments and Methods in Physics Research A 651 (2011) 1–5

Fig. 1. Distribution of advanced neutron imaging facilities around the world, according to data in Table 2.

Now need to add the ORNL HFIR (USA) to this list and the future SNS VENUS neutron imaging facility!

New Techniques

- The improvements in detectors has changed and impacted the neutron imaging techniques based on attenuation alone
  - The slow replacement of film methods by CR
  - The advances in neutron computed tomography
- The new methods referred to as phase contrast imaging take full advantage of recent facility, computer and detector advances
- Images with enhanced contrast are created by using the reflection, refraction, diffraction and ultra small angle scattering interactions

New Techniques

- Treimer (1996) at the Helmholtz Center in Berlin presented the first paper to demonstrate that the scattered neutron signal of weakly absorbing materials can be used to determine their location and size within a large matrix

- Treimer, et al. (1997) demonstrated the value of small angle scattering contrast imaging in NCT in the reconstruction of a bundle of 14-μm glass fibres

- Treimer, Ernst and Herzig (1997) reconstructed the magnetic field using polarized neutron imaging, and Kardjilov et al. (2008), published a paper on Three-Dimensional Imaging Of Magnetic Fields With Polarized Neutrons

- In 2000 a paper in Nature by Allman, et al., also demonstrated the contrast in low attenuating materials when using what they referred to as phase radiography with neutrons

Bragg Edges of Fe

- **Wavelength selective neutron imaging at a cold neutron reactor source was used to measure strain and determine (residual) stresses in a steel sample under plane stress conditions**

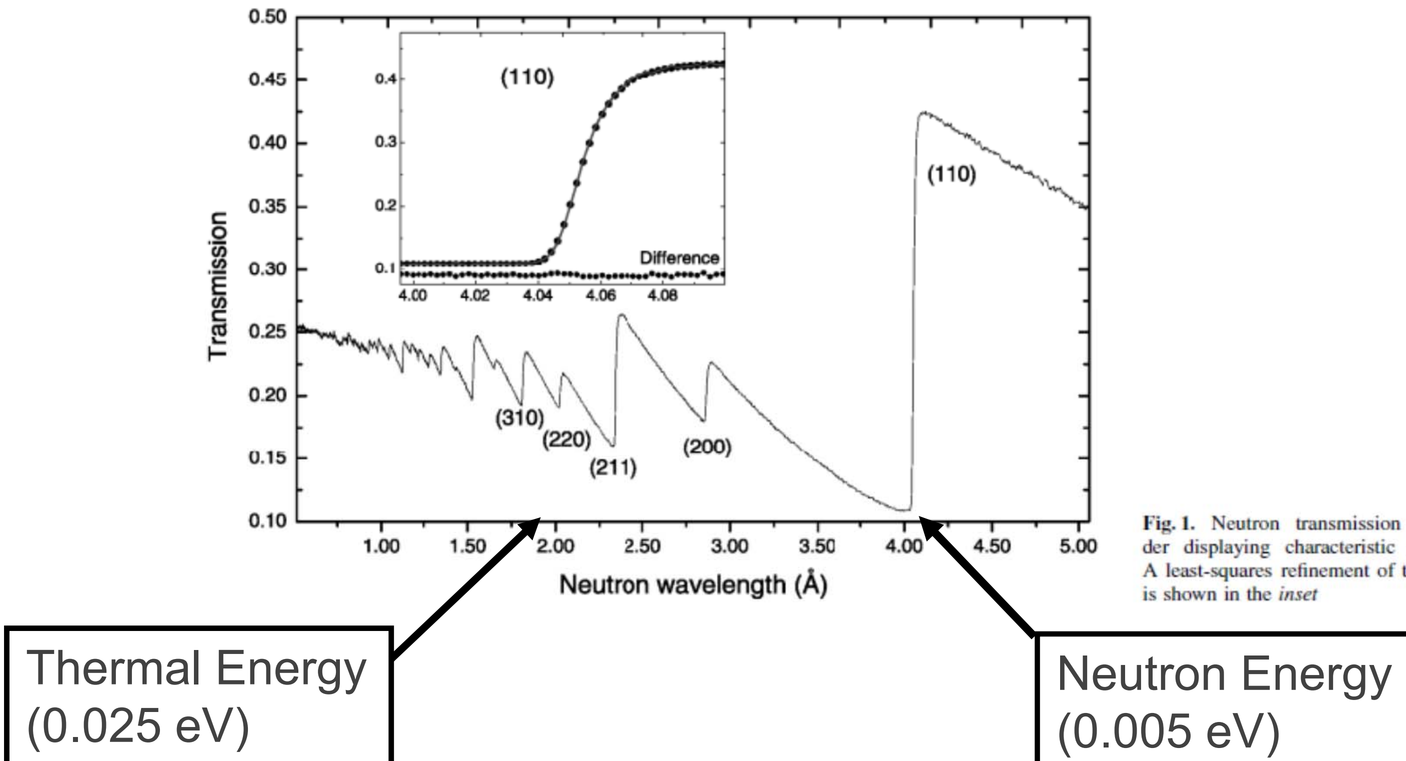

Fig. 1. Neutron transmission of Fe powder displaying characteristic Bragg edges. A least-squares refinement of the (110) edge is shown in the *inset*

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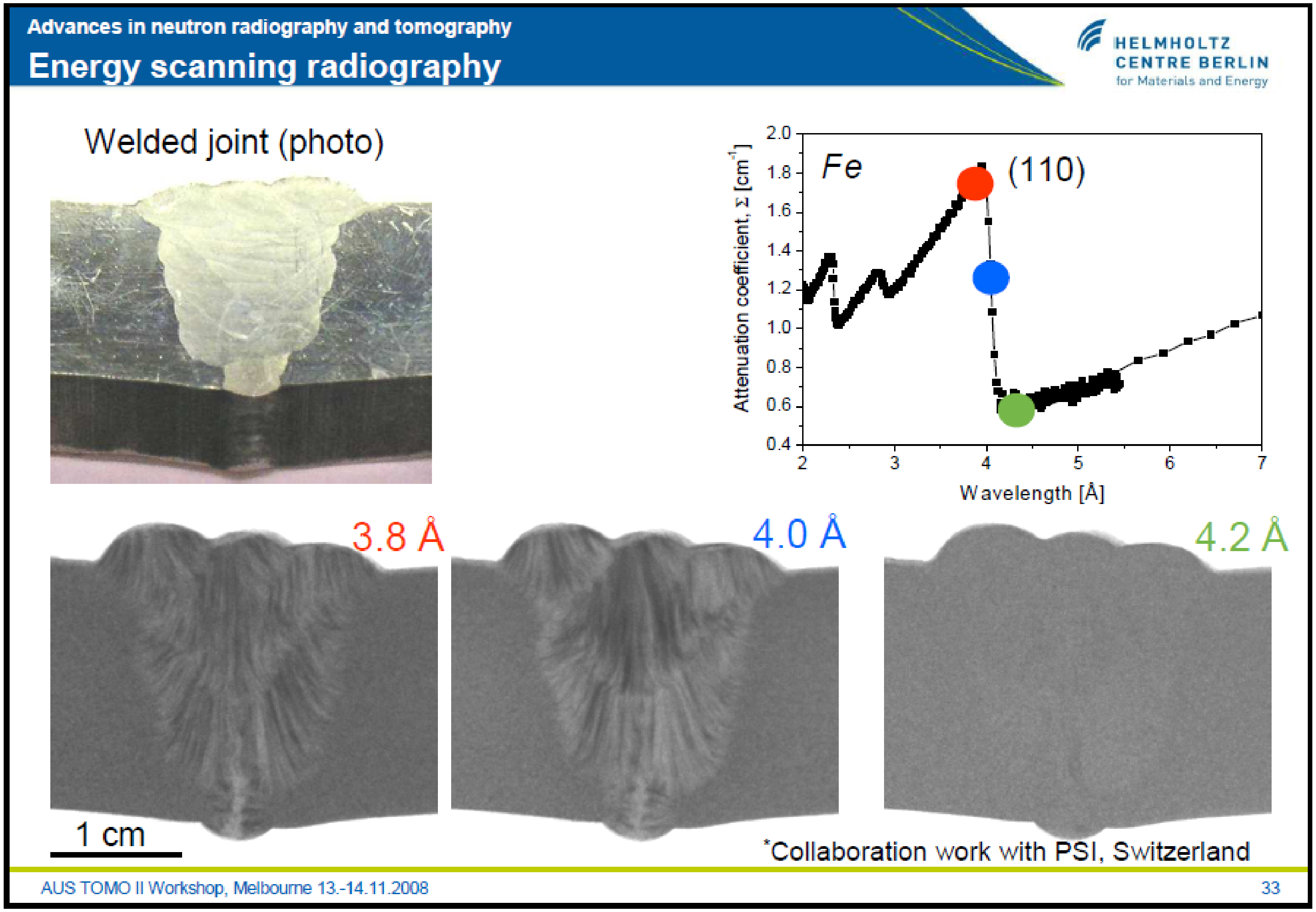
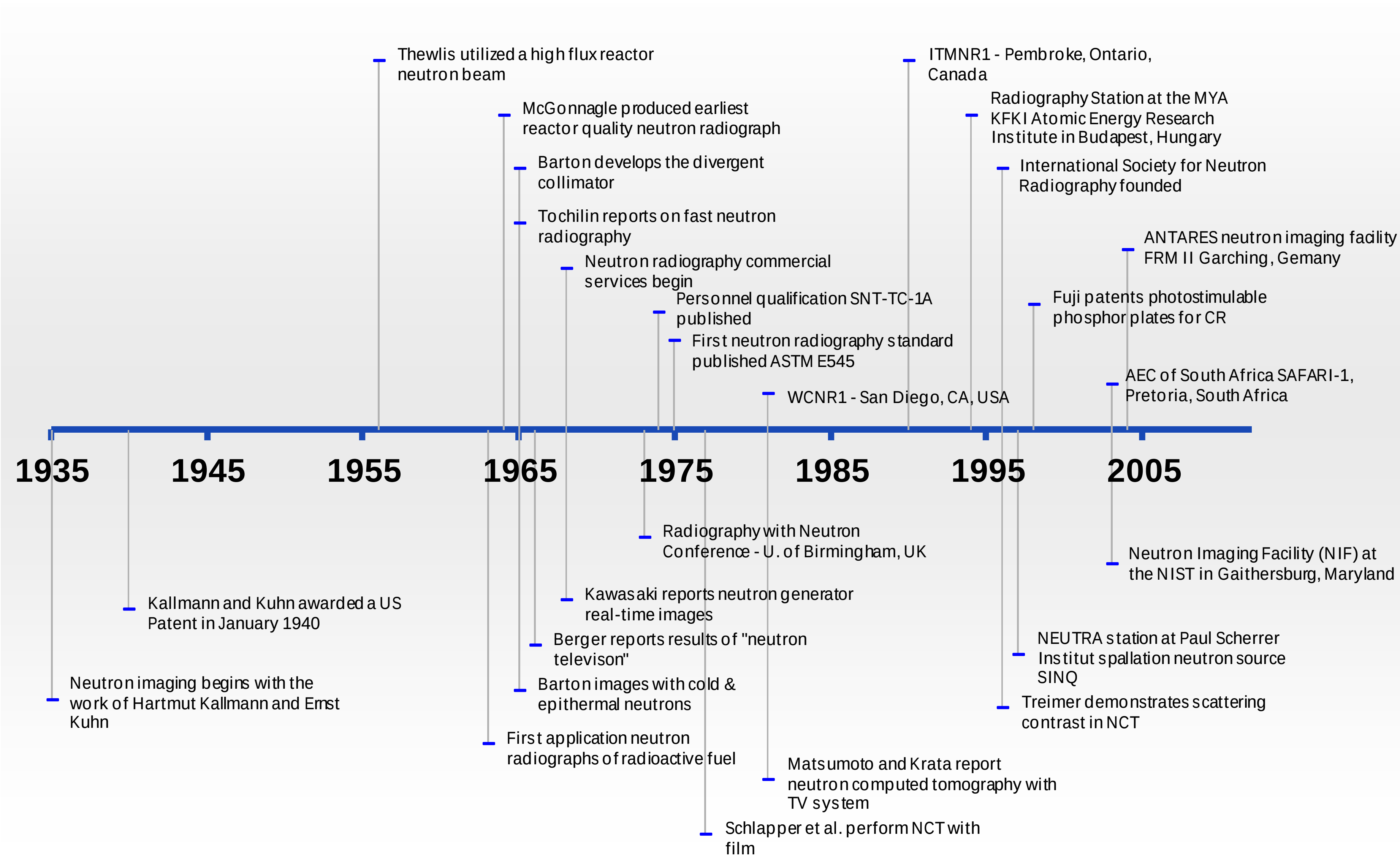
xrsi.cmit.csiro.au/TomoWkshop/PDFs/Banhart.pdf




Thewlis utilized a high flux reactor neutron beam
McGonnagle produced earliest reactor quality neutron radiograph
Barton develops the divergent collimator
Tochilin reports on fast neutron radiography
Neutron radiography commercial services begin
Personnel qualification SNT-TC-1A published
First neutron radiography standard published ASTM E545
WCNR1 - San Diego, CA, USA
ITMNR1 - Pembroke, Ontario, Canada
Radiography Station at the MYA KFKI Atomic Energy Research Institute in Budapest, Hungary
International Society for Neutron Radiography founded
ANTARES neutron imaging facility FRM II Garching, Gemany
Fuji patents photostimulable phosphor plates for CR
AEC of South Africa SAFARI-1, Pretoria, South Africa
1935
1945
1955
1965
1975
1985
1995
2005
Radiography with Neutron Conference - U. of Birmingham, UK
Neutron Imaging Facility (NIF) at the NIST in Gaithersburg, Maryland
Kallmann and Kuhn awarded a US Patent in January 1940
Kawasaki reports neutron generator real-time images
Berger reports results of "neutron televison"
NEUTRA station at Paul Scherrer Institut spallation neutron source SINQ
Neutron imaging begins with the work of Hartmut Kallmann and Ernst Kuhn
Barton images with cold & epithermal neutrons
Treimer demonstrates scattering contrast in NCT
First application neutron radiographs of radioactive fuel
Matsumoto and Krata report neutron computed tomography with TV system
Schlapper et al. perform NCT with film

Conclusions

- As neutron imaging approaches its eightieth year as a NDT method, it is important to keep in mind the seminal events that brought neutron imaging to its present state
- Quantitative measurement has become a very important and valuable NDT research tool
- The desire for higher resolution and contrast images (either analog or digitally reconstructed from the data) to observe smaller and smaller features
  - Challenges the current state-of-the-art detectors
  - Demands more intense neutron beams
- Interest imaging with neutrons continues to grow both in methods and in applications

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Questions?