

B. T. (Terry) M. Willis¹ – *A tribute*

Early years

Terry Willis obtained an honours degree in physics from the University of Cambridge in 1948, and a PhD from the University of London in 1951 working with Samuel Tolansky on the optical properties of piezoelectric crystals.

After his PhD in 1951 he joined the Research Laboratories, The General Electric Company, in Wembley, Middlesex where he worked with X-ray diffraction on different transition-metal compounds, and became a specialist at studying phase transitions as a function of temperature. His first paper (Willis & Rooksby 1952) was on haematite (α -Fe₂O₃), followed by one on hexagonal Mn compounds (Willis & Rooksby 1954).

While at GEC in Rooksby's X-ray analysis group he met George Bacon (1917 – 2011), who was visiting GEC. Bacon told Terry about Harwell and encouraged him to apply there. He was interviewed in 1953 and arrived at Harwell in early 1954.

At that time experiments were being conducted at the BEPO reactor (1948 – 1968), principally by George Bacon, Ray Lowde, and Bas Pease. One of their aims was to try and find the positions of the hydrogen atoms in KH₂PO₄, an activity also at two other neutron laboratories in the USA. Terry joined the Metallurgy Division [led by Peter Murray (1920 – 2011)] at Harwell, and began working with x-rays on radiation damage in LiF₄. He was interested in the possibility of using neutrons, but there were only a few ports at BEPO, so there was no room for any further instrumentation. However, the 26 MW reactors, DIDO (1956 – 1990) and its twin PLUTO (1957 – 1990), came on line in the late 1950s and provided space for a single-crystal diffractometer. With the support of the Metallurgy Division management, Terry started construction of such a machine to measure individual reflections from a single crystal. A second diffractometer was later built at the PLUTO reactor.

The diffractometer's design and automation were greatly aided by Terry meeting with Ulrich Arndt (1924 – 2006), who was then at the Royal Institution in London. They persuaded Ferranti to construct a new diffractometer, which operated with a punched paper tape and worked collecting intensities after 1960. This may have been the first commercially constructed diffractometer at a

¹ This article is based on an interview with Terry Willis at his home in Oxford on 9-July-2016. I am grateful for corrections made by Margaret and Terry Willis to the original account. Gerry Lander, Grenoble, France (October 2016)

neutron source, although one was also in operation at Oak Ridge being run by Levy, Busing, and Peterson.

In 1966 Arndt and Terry published a book “Single Crystal Diffractometry”. The book was a great success, and was reprinted in paperback in 2009.

Contact with Universities – widening interest in neutrons

Many different problems were tackled by the neutron diffractometer at DIDO and this led to Terry coming into contact with many academic staff at Universities. Certainly the most famous was Dorothy Hodgkin (1910 – 1994) at Oxford, who received the Nobel Prize in Chemistry in 1964 for solving, with x-rays, the crystal structure of vitamin B₁₂. She also had published the structure of insulin in 1955.

Hodgkin was most interested in using neutrons, especially to discriminate between carbon and hydrogen, and a number of crystals of inorganic materials were examined at Harwell. In the late 1960s early 1970s they focused on the corrin nucleus, which is in many biological systems, and consists of a small number of hydrogen, nitrogen, and carbon atoms. Of course, this work was slow, but it, together with the work of Benno Schoenborn at Brookhaven, were the first *single-crystal diffraction* experiments on biological macromolecules. Younger people such as Sax Mason (now at the ILL), Joe Zaccai (ILL, see his article on “Neutrons in Biology: Personal reminiscences of early days” on this website) and David Worcester (now at NIST) were also involved. The latter modified a so-called Badger diffractometer to be able to study reflections at low angles from membranes and measure small-angle scattering.

Although Terry and his collaborators were working at the reactor, they had little contact with many other scientists, for example Peter Egelstaff, Ray Lowde, Colin Windsor, Mike Hutchings, Bruce Forsyth, Cedric Turberfield, and Alan Wedgwood, who were then in the Materials Physics Division headed by Mick Lomer (1926 – 2013). Being in a different Division allowed Terry more freedom to pursue collaborations. Already in the mid 1960s, there had been encouragement for the scientists to work with outside users and the first real users programme was started at Harwell, with Peter Egelstaff (1925 – 2015) playing a crucial role. So Terry’s effort at broadening the base with neutrons was well received by Harwell management. Walter Marshall (1932 – 1996), the same Marshall of the famous textbook on neutron scattering Marshall & Lovesey, became Director of Harwell in 1968 and was charged with “making Harwell pay for itself”. This was a great challenge, and led to many different initiatives, some successful, some not. (A full account of this may be found in the book by Nick Hance, “Harwell – The enigma revealed”, 2006). Marshall supported Terry and

told him that 20% of Harwell's research should be basic; and Terry's programme was not affected by the changes at Harwell. This was not true for many others.

In the early 1970's the Science Research Council contributed directly to the cost of beamtime at Harwell for academic users through a formal agreement: the 'UKAEA-SRC Joint Programme'. In 1973 the UK joined the Institut Laue Langevin in Grenoble, and the Research Councils' funding for neutron scattering increased to pay the contribution to the ILL, and, shortly later, the planning of the new spallation source at ISIS. This ultimately led to the ending of the Joint Programme, although some university use of the beams continued through collaboration until the reactors shut down.

Terry's ongoing research in the 1960s and later on a variety of systems led to another book "Thermal vibrations in crystallography" together with Arthur Pryor, published in 1975.

Many scientists visited with Terry at Harwell in the 1960s and 1970s. Hugo Rietveld (Petten) came many summers and Terry and he discussed the profile fitting method that now bears Rietveld's name. Alberto Albinati came from Milan, and Noriaki Kato from Japan came a number of times. Terry himself was a visitor in Denmark, India, Switzerland, and Japan.

The saga of uranium dioxide and its defects

By the mid 1950s the development of nuclear power was being pursued in many countries, including the UK, and the limitations of uranium metal as a fuel were becoming evident. Although the metal has good thermal conductivity, methods of fabrication and the low melting temperature (1132 C) posed considerable problems. MAGNOX fuel, based on U metal, was developed in the UK, but more efforts were turning to the oxide, UO₂. The overwhelming advantages of ease of fabrication (using standard methods to produce ceramic pellets) and the high melting temperature (~ 2900 C) had significant implications for safety, so, despite its poor thermal conductivity, UO₂ gradually became the fuel of choice, as it still is today. Of course, UO₂ is a perfect system to study with neutrons as the x-ray pattern is dominated by the scattering from uranium, and by 1961 single crystals were available from a number of sources, including some that were grown at Harwell. Terry thus started on a trail that was to last more than 50 years!

Terry published the first structural work on stoichiometric UO₂ in 1963 (Willis 1963a and b). In 1963b they reported measurements up to 1100 C, and showed that at these high temperatures normally equivalent reflections had different intensities. Although initially these differences were ascribed to shifts in the

oxygen atom positions from their special positions in the $Fm\bar{3}m$ CaF_2 structure, they later realized (Rouse et al, 1968), following the theoretical work of Dawson 1967, that the correct description was that the vibrations of the oxygen atoms are *anharmonic*. In fact, this is a general property of the fluorite structure where the anion point symmetry is $\bar{4}3m$. The agreement between experiment and theory was startling.

Already by the late 1950s it was known that the U-O phase diagram was complex, with at least three phases UO_2 , U_4O_9 , and U_3O_8 , with different phases of U_4O_9 found as a function of temperature. The fission process generates much heat and also defects in the UO_2 structure – the crucial question became what happened to the UO_2 lattice as oxygen was absorbed into the lattice? This was addressed by Terry in a seminal paper in 1963 on a single-crystal sample of $UO_{2.13}$ measured at 800 C (Willis, 1963c). The answer that Terry proposed was that two vacancies were formed together with the occupation of two interstitial sites, one along $\langle 110 \rangle$ and one along $\langle 111 \rangle$ away from the position of the oxygen vacancy in the fluorite structure. Taking the equivalent positions of the interstitials this gives four oxygen interstitials in place of the two vacancies, so allowing the oxygen content to rise above 2.00. The resulting cluster is called the “Willis 2:2:2 cluster” and is widely cited. Of course, over the years this model has been refined, Willis 1978 and Murray and Willis 1990, but the essential details have not changed since the first paper in 1963 (Willis 1963c), and the longer paper soon after, Willis 1964.

The importance of these concepts was underlined in a recent paper by Wang, Ewing, and Becker in 2014, where they presented model calculations of the position of the interstitials in the fluorite structure, and claim that “*We demonstrate that the Willis cluster is a fair representation of the numerical ratio of different interstitial O atoms; however, the model does not represent the actual local configuration. The simulations show that the average structure of UO_{2+x} involves a combination of defect structures including split di-interstitial, di-interstitial, mono-interstitial, and the Willis cluster, and the latter is a transition state that provides for the fast diffusion of the defect cluster.*”

The Willis 2:2:2 cluster is shown in Fig. 1, which is taken from the work of Wang *et al*, 2014.

Understanding the defect configurations is important for safety considerations. Experiments have shown that, except at very high burn-up, the clusters do not migrate in irradiated UO_2 , but are relatively stable and just increase in density as a function of burn-up. The integrity of fuel is maintained under irradiation.

Terry also tackled the crystal structure of U_4O_9 , which had first been identified by x-rays in the late 1940s. Single crystals of this material were prepared directly by controlled oxidation of UO_2 single crystals. The first paper appeared in 1986 (Bevan *et al.* 1986) with further details appearing in Cooper and Willis 2004. In an article by Garrido *et al.* 2006, Terry was pleased to correct a paper using EXAFS, which unfortunately involved the present author, purporting to show that there was a short U–O distance of $< 2 \text{ \AA}$. This showed Terry’s combative spirit even in his 80s, and the present author has no defense to offer!

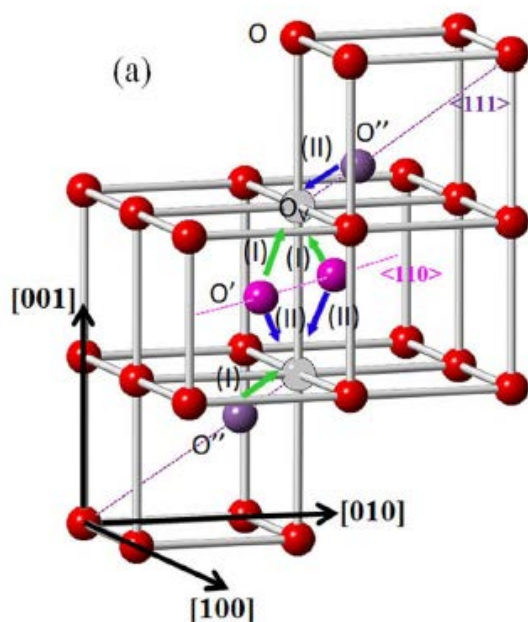


Fig. 1
Schematic structure of the Willis 2:2:2 cluster. The arrows indicate the movement of the atoms around the center vacancies. Spheres are oxygen atoms; uranium atoms are not shown. Red are lattice 8c O atoms, purple and pink for O interstitials displaced along $\langle 111 \rangle$ and $\langle 110 \rangle$, respectively. Grey circles are the oxygen vacancies at the 8c site.
[Taken from Wang *et al.* (2014)]

To close the UO_2 story, its magnetism is worthy of mention. UO_2 had first been demonstrated magnetic by the specific heat measurements of Jones *et al.* 1952, which showed a spectacular first-order phase transition at $\sim 31 \text{ K}$ that suggested antiferromagnetic ordering. Henshaw & Brockhouse at Chalk River National Laboratory in Canada reported the first neutron measurements on UO_2 in 1957. Terry had been involved in low-temperature work in the early 1960s, but was so busy with the structural work that the study of the antiferromagnetism lay unpublished in his office. However, in late 1964 he made a visit to Brookhaven National Laboratory on Long Island and found, to his surprise, that they were working hard on the study of antiferromagnetism in UO_2 . He returned to the UK and quickly wrote up what they had at Harwell and submitted it (Willis & Taylor 1965) two weeks before the longer BNL paper by Frazer *et al.* 1965 was sent to Physical Review. Of course, the story of the magnetism of UO_2 went on and on, both elastic and inelastic scattering, and came to a (perhaps only temporary) end with the review article of Santini *et al.* (2009), and the report of excellent agreement between theory and experiment for the inelastic studies by Caciuffo *et al.* (2011). The magnetic story has been as long and twisted as the structural one that Terry started in 1963, but was not centered at Harwell.

The Oxford Schools on Neutron Scattering

Although the uranium dioxide story did not involve any academics, at least in the early stages, Terry was very much involved with visitors coming to Harwell when he had his diffractometer working in the early 1960s. It was therefore natural for him to be approached by Mick Lomer in 1965 about organizing a “School for Neutron Scattering”. The first school was held at Harwell in 1966 and subsequent schools were held every alternate year until 1974, then there was a pause when Terry was not well², and the schools started again in 1979, but were not on a regular basis until the mid-1980s, when they were moved to Oxford, and continue on a biennial basis. We are now looking forward to the 16th Oxford Neutron Summer School in September-2017. As far as we know, the school in 1966 was the first one in the world, and many famous neutron practitioners have attended the schools of the past 50 years, and hopefully it will run for many more.

Many pictures of the school participants can be found on

<http://www.oxfordneutronschool.org/history.htm>

Past attendees are encouraged to identify themselves to osns@stfc.ac.uk if you find yourself unidentified in a photo.

Gordon Squires (1924 – 2010) from Cambridge gave the theory section at the first few schools. This was later given by Mike Gunn (University of Birmingham), and more recently by Andrew Boothroyd of Oxford. Gordon Squires turned his lectures into a book (Squires 1978) that was (and still is) enormously popular for those starting in the field, and it has been re-printed a number of times.

Colin Carlile helped Terry re-start the schools, and their efforts for the school also turned into a book (Willis & Carlile, 2009).

Just one story on the re-start of the schools in the 1980s. Terry left Harwell in 1984 for a position with the Chemical Crystallography Laboratory at Oxford University. However, the University was not keen to subsidize the school, as it did not see supporting neutron scattering as a priority. At that time Terry and Colin had arranged to have the school participants stay in Mansfield College in Oxford (attendees have more recently lodged in St. Anne’s College), and Mansfield demanded a deposit of £15,000 against the possibility that no students would turn up! Terry paid this from his own savings. Fortunately, the students

² Terry apparently became indispensable to the school. If he didn’t organize it, nobody did. The British were still trying to get their own reactor in the early 1970s – see *Nature* **239**, 60 (1972) but the powers that were (see article) preferred to spend their time politicking rather than organizing a school to bring in more users!

did indeed come, but the school had difficulties of this sort until it was taken over some years later by the Rutherford Laboratory, and since then has flourished without anybody's savings been depleted!

Many of those who attended the early schools, or played a role in their organization, attended a Celebration at Cosener's House in May 2009. The photo and list of participants is below.



Neutron Summer School Celebration – 21 May 2009

Back row from left to right

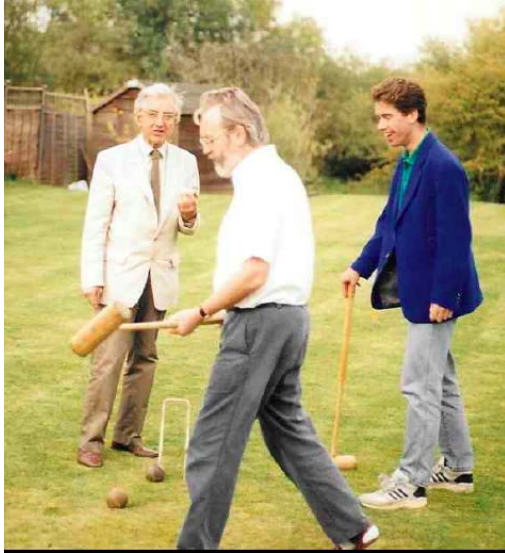
Alan Leadbetter, Mike Johnson, Brian Rainford, Richard Nelmes, George Stirling, John Dore, Roger Stewart, Brian Fender, Gerry Lander

Middle row from left to right

Ray Fenn, Mike Hutchings, Andrew Taylor, Hartmut Fuess, Mogens Lehmann, Peter Webster, John Tomkinson, Martyn Cooper, Adrian Wright, David Howard, Keith Ross

Front row from left to right

Noor Butt, Shelia Hutchings, Brenda Leadbetter, Gordon Squires, Ruth Fenn, Colin Carlile, Terry Willis, Judith Howard, Alberto Albinati



A tradition in the early schools was “Tea & Croquet” at the Willis’s in Oxford on Saturday afternoon. The picture to the left, from the 1993 school, shows Colin Carlile just finishing a shot, and Terry (replete with tie and jacket) pronouncing the probable bad news that Colin has not done what he should have done for his team.

In 2007 the Neutron Scattering Group of the Royal Society of Chemistry established a prize for outstanding neutron scattering science. The prize is named in honour of the founding chairman of the Neutron Scattering Group, B T M Willis and is awarded annually.

Conclusions

Terry has had a remarkable career, as a pioneer in neutron crystallography at Harwell, as one of those who established the first User Programme, and then the first school on the subject, and made seminal contributions with his work on uranium dioxide. It is fitting to finish this tribute with a poem written for the 2009 celebration by Alan Leadbetter (former head of ISIS and Scientific Director of the ILL), which is attached below.

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I'm going to tell you an interesting story
That relates to most everyone here
It started way, way back in sixty six
At least I think that that's the right year!

It was in a place you all know called Harwell,
Where B T M Willis could be found-
Scattering neutrons from all kinds of crystals
And look - the old chap's still around!

Well, he was a great expert on neutrons
And wished to spread his know-how around
So he thought of a School for Neutrons (not
scandal!)
And the idea fell on fertile ground.

Oh, it was such a splendid time to be living,
Life was so exciting, fulfilling and free
You could do whatever research that you wanted,
And not word had been heard of R A E.

But most of us had never met a neutron,
Didn't even know what they did,
Let alone how to find and then use them-
'Til the good men of Harwell raised the lid.

Their teamwork was really tremendous
When they invited us onto the site
To learn all about using those neutrons,
Then ensuring we did everything right.

But most of us were pretty darned ignorant
And our students the same - but much more
So some really good training was needed
And this brought young Terry to the fore.

The Harwell Neutron Schools were started
And it's now plain for the whole world to see
What a great contribution they've made
In bringing us right to the top of the tree.

So Terry, we congratulate and we thank you
For your achievements over all of these years
With your excellence, good humour - and stamina
You are honoured among all of your peers

Now my final remark of the evening
In keeping things brief - as I should-
Is that it's clear when I look at you oldies
That **Neutrons - they do you Good !**

So I now propose a toast to Terry-
Please be upstanding and raise your glass
And we'll drink to his health and his happiness,
This gentleman of science and rare class!

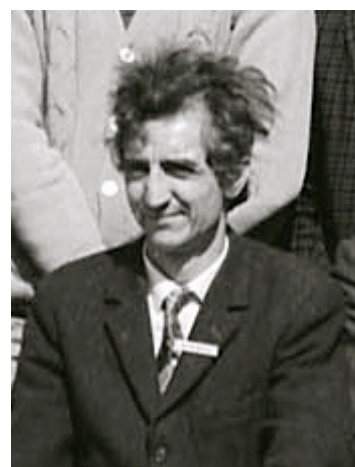
Alan Leadbetter
21 May 2009



1966



1968



1970