The strange history of polarised neutrons in Australia

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Abstract. The history of polarised neutrons in Australia is unusual firstly because of the particular access that individuals in universities had to the facilities at the reactor site and because this resulted in the experiments being done almost all being with polarisation analysis. Two instruments were initially available. One was a conventional instrument albeit with a tilting counter. The other was a primitive polarisation analysis instrument purpose built for diffuse scattering. This latter instrument evolved over more than thirty years and produced results ranging from the separation of magnetic and nuclear diffuse scattering, for which it was conceived, to the isolation of magnetic features in inelastic spectra.

1. Introduction

The first polarised neutron experiments at the HIFAR reactor in Sydney were unusual in several respects. The first was in the method of accessing the facilities on the reactor site and the second was that the experiment was a polarisation analysis one.

In 1958 the Australian Institute of Nuclear Science and Engineering (AINSE) was formed. This was to be a bridge between the Australian Atomic Energy Commission (AAEC) and the universities. The then chairman of the AAEC set out the philosophy at the inauguration of AINSE. He said

"Not even the wealthiest university can equip itself completely for the study of nuclear energy today. The universities provide basic training in nuclear science and engineering with their own equipment and staff, but in any advanced programme they have to access to a nuclear reactor and specialised chemical, engineering and metallurgical laboratories to handle highly radioactive materials." J P Baxter.

The role of AINSE was: To provide the universities access to the facilities of the AAEC and to provide staff and students of those travel and accommodation to access those facilities. The AAEC and the universities provided funds and this resulted in Australia having the world’s first user programme for neutron beams among other facilities. Proposals were sought and peer reviewed and accepted or rejected.

It should be emphasised that this user programme was for individuals within universities and not for the universities themselves. The result was that the university or AAEC bureaucracies had no influence on the success or failure of particular proposals.

In the following sections we will outline how it was that the first polarised neutron experiment was a polarisation analysis experiment rather than a simple polarised beam one.

2. First polarised neutron instrument

By the late 1960s more instruments and support were required to satisfy demand than could be provided by the AAEC. AINSE then formed its own neutron diffraction group consisting of Frank
Moore, David Wheeler and Roy Ebdon. One of their first projects was to build a versatile polarized neutron diffractometer.

At that time there was a potential demand for such an instrument from Brian Figgis, a computational chemist from the University of Western Australia. So the instrument was built by the AINSE group. It was a classic instrument with a CoFe monochromator polarizer. It had a vertical field at the specimen position and a tilting counter to access other reflection zones. Figure 1 shows the instrument in 1972 [1]

![Figure 1](image.png)

Figure 1. The first polarised neutron diffractometer at Lucas Heights. Note tilting detector. The photo was taken in 1972.

As it happened Brian Figgis never used this instrument. Instead he collaborated with colleagues from ILL.

3. **LONGPOL I**

At almost the same time another polarised neutron instrument was being constructed. This resulted from a proposal to AINSE to isolate the magnetic diffuse scattering in the dilute alloy systems Cu-Mn and Cu-Fe. Cu-Mn was widely regarded as a spin glass and it was known that that there was cusp in the magnetic susceptibility but it was not at all clear whether this was due to some degree of magnetic ordering or a simple freezing.

The new instrument was based on a previously existing helical velocity selector with out of pile shielding and a wavelength resolution of 10%. The diffractometer itself was single axis and the detector was heavily shielded and rested on airpads on the floor. The detector was rotated by manually pushing it while the airpads were pressurised. Polarisation and analysis were achieved by polycrystalline iron filters in electromagnets before and after the specimen. It was important that the filters were magnetically saturated. The result was that the Bragg scattering in the filters was different for up and down neutrons which discrimination peaking at the Bragg cutoff. Immediately below the Bragg cutoff (3.6 Å) the cross sections for up and down spin neutrons were in a ratio 2:1 which for comparison is equivalent to a He$^3$ filter with a 65% He$^3$ nuclear polarisation. As for He$^3$ polarisers the
thickness of the filters is a compromise between perfect transmission and perfect polarisation. The iron filters have broad angular acceptance and relatively broad wavelength acceptance which is very good for a diffuse scattering instrument, like LONGPOL, not requiring good resolution. Stewart Campbell, Naeem Ahmed and I surveyed many ferromagnetic and ferrimagnetic materials by calculating their up and down neutron spin cross sections but we found none better than iron [2,3]. Finally LONGPOL had an RF flipper mounted before the specimen so that the incident beam could be spin flipped. To separate magnetic from nuclear scattering the polarisation was directed along the scattering vector at the specimen.

Figure 2 shows the instrument with Naeem Ahmed behind

![Figure 2](image)

Figure 2. The first LONGPOL in 1972. The detector was positioned manually on the airpads.

LONGPOL I provided the first direct evidence that Cu-Mn was a spin glass. A cusp in the variation with temperature had been observed in the magnetic susceptibility. However this was similar to that observed for the antiferromagnetic critical temperature. No long range order had been observed and it had been surmised that the magnetic distribution was glass like. LONGPOL was able to directly measure the magnetic diffuse scattering [4] and separate it from the scattering from the nuclei and the results are shown in figure 3.
Figure 3. Diffuse scattering from Cu 5 at% Mn. (a) Nuclear cross section. (b) Magnetic scattering at various temperatures. Susceptibility values are also shown for the various temperatures. A typical error bar is shown.

Plot (a) shows the nuclear scattering and plot (b) shows the magnetic scattering. The nuclear scattering is flat as function of scattering vector indicating that there is a random distribution of Cu and Mn nuclei, but there is some correlation in the magnetic scattering which develops slightly with lower temperature.

Furthermore the small scattering vector cross section of the magnetic scattering was compared with the bulk susceptibility. The cross sections were placed on an absolute basis and the cross sections derived from the susceptibility plotted at zero scattering vector for each of the scans taken. It can be seen that at the two lower temperatures any extrapolation to zero scattering vector of the scattering will not match the susceptibility measurements. At the two higher temperatures extrapolation of the scattering does match the susceptibility. This means that at high temperatures the magnetic fluctuations are free to respond to a magnetic field. At lower temperatures a large fraction of the magnetic fluctuations are too slow to respond to a magnetic field, that is, they are frozen or glass like. Further Cu-Mn experiments and experiments on similar alloy systems were also performed at this time.
4. LONGPOL II

LONGPOL was shifted to a more intense beam and the wavelength selection was changed from a velocity selector to a graphite double monochromator. Apart from this the basic diffractometer remained the same. Figure 4 shows the modified instrument.

![LONGPOL II](image)

The out of pile shielding was retrieved from the AAEC’s junkyard. The RF flipper and cryostat are shown more clearly in this photo.

The other major modification was to select elastic scattering. To achieve this the RF flipper was pulsed randomly and the detector was gated with the same pulse sequence delayed at the neutrom time of flight. This was implemented by John Davis. Just to confirm that it was effective, a magnon in antiferromagnetic Mn-Cu was measured in neutron energy gain by altering the delay [5].

The investigation of magnetic diffuse scattering was extended to antiferromagnets at this time. An example is that from a single crystal of antiferromagnetic Mn-Ni alloy as seen in figure 5. The left panel shows the separated nuclear scattering from the crystal along these directions. Clearly the distribution of atoms is not random but the distribution statistics can be obtained from these measurements. The right panel shows the magnetic diffuse scattering which reflects both the distribution of magnetic atoms and the variation of magnetic moment due to their environment [6]. The striking feature is the peak in diffuse scattering which occurs near the 001 position. Assuming the antiferromagnetic structure is collinear the absence of a Bragg peak at 001 indicates that the spin direction of the structure is along z. The magnetic fluctuations at this point must therefore be transverse in order to contribute to the diffuse scattering.
Figure 4. Diffuse scattering from antiferromagnetic single crystal Mn Ni. On the left is the nuclear scattering. The magnetic scattering is on the right. (a) 001 direction. (b) 110 (c) 111.

LONGPOL III
The makeover of LONGPOL at this point was an attempt, after fifteen years, to engineer the instrument. A further move to a yet more intense beam was made however with a more than ideal length of throw between monochromators due to the constraints of the reactor beam hall. Figure 5 shows the layout of the new instrument.

Figure 5. Layout of LONGPOL III
From the double graphite monochromator the beam passes from the out of pile shielding into the instrument’s barrel shielding and through the higher order filters. The beam is the polarized by an iron filter and flipped by a Mezei coil before passing through the specimen position with the ability to turn the polarisation horizontal. The scattered beam is analysed by a magnetized iron strip before being detected by one of the eight detectors. Barry Maguire developed the software control of the instrument. The polarizing and analyzing filters would later be replaced by supermirrors. The building of the instrument was a team effort. The AAEC (Ansto) provided the out of pile shielding and the diffractometer table with the detector arm. AINSE provided the barrel shielding and Monash University the analyser magnet with the Helmholtz coils which provided the background field and which was financed by national competitive grants. The instrument with the iron filters is shown in figure 6 with Shane Kennedy looking on.

Figure 6. LONGPOL III with Shane Kennedy. The edge of the electromagnet holding the analyzing iron filter can just be seen.

At this point it is appropriate to tell the story of the implementation of supermirror polarisers. A supermirror polarizer was first fabricated in 1982. Shane Kennedy while a research assistant at Monash University used an old evaporator to deposit layers of Fe-Co and Al on a silicon wafer which provided good reflectivity and polarization out to $2\theta_c$. Fifteen years later we were able to arrange to
have similar supermirrors sputtered at HMI for incorporation into stacks for the detectors and for a polarizer. A stack is shown in figure 7

Figure 7. Compact supermirror assembly. The stack is held in the red permanent magnet. In the front is the cam to adjust the silicon stack curvature.

The permanent magnet provided the field to magnetise the layers and a cylindrical cam provided the ability to adjust the curvature of the wafers. Stack dimensions were 7x5x2 cm. Figure 8 summarises the stack performance.

Figure 8. Performance of a supermirror bender stack.
Basically the stacks had 57% transmission of the right spin, close to 0% transmission of the wrong spin with 97% polarization over 20_c with an acceptance angle of 1° at 3.6 Å [7].

There were many diffuse scattering experiments performed with this instrument but some results highlighting the wide coverage of phase space in the investigation of spin orientation in a mixed ferromagnetic antiferromagnetic structure. Figures show the peaks in ferromagnetic Fe_2MnSi from the antiferromagnetic modulation. Lack of a 111 means that the antiferromagnetic direction is <111> and because the <113> is completely spin flip the antiferromagnetic modulation is collinear [8].

![Figure 9. Bragg scattering from antiferromagnetically modulated ferromagnetic Fe_2MnSi. Spin flip scattering on the left. Non spin flip scattering on the right.](image)

With increased intensity and multidetectors some time of flight experiments were attempted with pseudo random pulsing of the flipper. Figure 11 shows a spectrum of crystal field excitations in PrAl3 taken with the iron filters still in place. With the neutron spin along the scattering vector all magnetic features are with spin flip whereas the features with no spin flip are from nuclear scattering. Consequently the magnetic features in the spectrum are negative going and the nuclear features are positive going.
Figure 10. Spectrum of PrAl₃. Positive going features are magnetic. Negative going features are nuclear.

The spectrum shows the nuclear elastic peak at 6.0 meV against a magnetic quasielastic peak. At a neutron energy of 10.5 meV there are two barely resolved crystal field excitations [9].

Figure 11. Magnetic inelastic scattering from CeCu₆ at 25 K.

Figure 12. Magnetic inelastic scattering from CeCu₆ at 50 K.
Later a more serious attempt was made to separate out the crystal field spectrum from CeCu₆. Spectra were taken with the neutron spin along the scattering vector and at right angles perpendicular to the scattering plane and the difference taken. This should, apart from nuclear spin scattering, be only magnetic scattering. The error bars were obtained by collecting many data sets for each temperature and for each neutron spin orientation, cross correlating each, averaging and taking the standard deviation at each point.

The spectra show quasielastic scattering at each temperature but broader at 50 K. There is also an excitation at 7 meV at 25 K which is rather washed out at 50 K. These are consistent with other unpolarised studies but now these features are clearly identified as magnetic [10].

**Conclusion and present day**

From 1971-2, when the two instruments incorporating polarised neutrons were first built at the HIFAR reactor there has been a continuous polarised neutron programme at Lucas Heights only interrupted by final shut down of HIFAR. With the advent of the OPAL reactor there has been an intention to provide polarised neutrons on a wide range of instruments. At time of writing there are polarised neutrons on several instruments.

There is currently a programme to provide He³ filters for wide use but the most ambitious project is to provide analysis covering the whole detector bank of the cold neutron time of flight instrument PELICAN (son of LONGPOL). When this is complete PELICAN will be able to do many of the experiments attempted on LONGPOL but with orders of magnitude more intensity.

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