

## FUNDAMENTAL MATTER \*

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*This neutron-scattering study deals with the behaviour of the order parameters  $M_{st//}(H,T)$  and  $M_{st\perp}(H,T)$  in  $\text{CsMnBr}_3 \cdot 2\text{D}_2\text{O}$ . The data provide a first direct test of the extended-scaling theory near the critical point of a spin-flop system.*

What is stated here? The authors of this summary, Evert Frikkee, Jan Basten and Wim de Jonge, measured the neutron scattering by an anisotropic anti-ferromagnet, and found that the order in this magnet, both parallel and perpendicular to the natural spin-direction, does not only depend on the temperature but also on an external field. For the first time they showed in a direct way that Wilson's theory on the universal character of critical behaviour is also valid for anti-ferromagnets in which the spins can suddenly be directed in, or flip over to, a perpendicular direction.

So, in this way, by quoting this summary, we want to stress the scientific value of the research at the High Flux Reactor in Petten. The finding of the three scientists was fundamental, timely and beautiful. They published their result in *Physical Review Letters* **42** (1979) 897, whereas shortly afterward, in 1982, Kenneth Wilson got a Nobel Prize for his theory – which was quickly, only eleven years after its publication in *Physical Review* **B4** (1971) 3184. 'Petten' therefore played a role in the solution of an outstanding scientific problem, namely that of the forces behind the critical phenomena.

In the *sixties* and *seventies* many scientists were studying this question, effectively working together in a research programme with 'positive heuristic'. [The latter characteristic is due to Imre Lakatos, a philosopher who wrote in these years about the methodology of scientific research programmes.] The 'positive heuristic' meant that they tried to falsify renormalisation theory – the name used by Wilson himself – which predicted that the critical exponent  $\beta$  was 0.325. In the classical theory for critical phenomena, where the forces between the spins [or particles] were assumed to have an average strength,  $\beta$  had to be 0.5, but in reality it was lower than that: somewhere between 0.25 and 0.41. Frikkee and his colleagues obtained  $\beta = 0.321$  or  $\beta = 0.326$  – the first value for the case that the magnetic field was parallel to the spin direction, and the second for the case it was perpendicular to it. The difference with 0.325 was 1% or less. So, it was a brilliant confirmation of Wilson's prediction.

How could 'Petten' get a name in fundamental research like this? Without any doubt by the drive of Jaap Goedkoop, to whom the direction of the scientific use of the High Flux Reactor was entrusted.

Goedkoop: again his name is dropped. He needs a lot of our attention, not mentioning him only in passing [like we did to characterize his management of the Centre] but because of his direct involvement in the scientific work at the reactor.

He had pioneered in neutron scattering and showed what one could learn about vibrations in a crystal. In 1960 he discovered phonons in metallic hydrides, together with Jitze Bergsma, and spin waves in an anti-ferromagnet, together with Tormod Riste, using neutrons from the Experimental Pile in Kjeller, Norway. Being so quick with these discoveries, they were mentioned at once in [the second edition of] Bacon's great book on neutron diffraction – Goedkoop's name is mentioned there 4 times. It was also in Kjeller that the first world congress about nuclear energy had been held, already before the famous Geneva-conference of 1955. In that year Goedkoop worked for the United Nations and witnessed the disclosure of secrets about nuclear reactions and the relevant processing of materials, which were most interesting, both scientifically and politically.

'If I ever had underestimated the importance of the political side of nuclear energy,' he said, 'it was there and then that I lost this naivety. I am not naive. How could I have been director for so many years if I were? By the way, I don't know scientists who are.'

Isn't the scientific thesis the purest image of a man? Jaap's thesis is from 1952, when he still was 30. It is about X-ray diffraction by crystals, and filled with elementary mathematics to derive the probable value of crystal parameters – precise in wording and calculation, but original? 'Although written in Amsterdam,' the foreword states, 'it is largely based on measurements during a work placement in the laboratory of professor Pepinsky at the Pennsylvania State College in 1949–1952.' And the last words of the thesis are: 'A country like the Netherlands, being so dependent on shipping and industry, should much increase its funding of the development of nuclear energy.'

Let's listen to Bergsma who was mentioned before. We spoke with him in October 1998. If Jaap Goedkoop has accomplished one thing, he said, then it is the research programme for the neutron beams of the High Flux Reactor. The reactor in its bare form had ten radiation tubes, ending in cubical spaces where – so was the idea – materials could be irradiated. But for such irradiations one could better use sites in or close to the reactor core, where the neutron flux was higher. Moreover, the cubical spaces were too narrow for controlled scattering experiments, and 'poisoned' by background radiation. So, in 1960 Jaap went to Oak Ridge, where a similar reactor was in use, to see what one had done to make scattering experiments possible at the end of the radiation tubes. Back in Petten, he immediately ordered similar blocks of heavy concrete, in which background radiation could be absorbed, and narrow channels were left open for collimators and spaces for monochromator crystals. In 1963, four of these blocks were installed, and the next year two more. He allotted beams 1, 3 and 5 for Petten's own programme of solid state physics, and beams 2 and 4 for studies of nuclear physics by the Dutch Organization for Fundamental Research of Matter (ZWO).

The five beams, prepared in this way, were speedily provided with instruments. The flux at their exits was so high – up to a billion neutrons per square centimeter per second – that unique experiments could be done, i.e. unique for the time. The ZWO-members Ger van Middelkoop and Hans Postma measured  $(n,\gamma)$ -reactions in beams 2 and 4, while Kees Abrahams did the same in a programme of Petten alone. Neutron spectrometers for beams 1, 3 and 5 were built by Bert Loopstra, who was assisted by Bob van Laar for the diffraction, Evert Frikkee for the magnetic scattering and Jitze Bergsma for the 'common' inelastic scattering. In the *seventies* the beams 7 and 9 were added to the experimental programme. Kees Abrahams looked at nuclear reactions with polarized neutrons, whereas Bob van Laar, Hugo Rietveld and Rob Helmholdt, using beam 9, looked with high resolution to the magnetic diffraction by solids. In the *eighties* Cor van Dijk built a diffractometer for small-angle scattering in beam 5, while new apparatus was installed in beam 4 for the assesment of residual strain in metals. The latter adaptations were required by a new programme of applied research. Fundamental research was better served then by Harwell's spallation source, of which the neutron flux was two orders of magnitude higher than of the High Flux Reactor.

Bergsma, who told us the above, stressed that Goedkoop's role was just to initiate: 'to sow, not to harvest.' He didn't request to be added to the list of authors of papers based on experiments at the nuclear reactor. Loopstra, who had studied with him in Amsterdam and had come to Petten as well, was left completely free in his research. 'It was a pity that they didn't like each other, the formal boss and the free-thinker.' When Loopstra left in 1972 to succeed Caroline MacGillavry [who had been the thesis adviser of both of them], they didn't cooperate any more in the use of the beams. Goedkoop,

being extraordinary professor in Leiden and acquainted to colleagues in Amsterdam, Delft, Eindhoven, Enschede, Groningen and Utrecht, was sole promotor only for Bergsma and Van Dijk –

J. Bergsma, *Lattice dynamics of magnesium stannide and zinc blende*, Leiden 1970.

C. van Dijk, *Investigation of lattice dynamics of  $\alpha$ -Fe and Fe<sub>3</sub>Al by neutron inelastic scattering*, Leiden 1970.

And the other subjects?

Bert Loopstra was the first to understand the structure of the mixed oxide U<sub>3</sub>O<sub>8</sub> [from which the UO<sub>2</sub> fuel for nuclear reactors is derived], as he understood it to be 2UO<sub>3</sub> & UO<sub>2</sub>. This also was the first scientific finding at the High Flux Reactor.

Hugo Rietveld was the first to use a computer for the analysis of peaks in the diffraction pattern of powders studied in beam 5. Guided by the idea of Bert Loopstra and Bob van Laar that all peaks would have the same profile, he wrote a numerical code which dissolved the noisy pattern as it was measured in a number of peaks with that same profile. His code worked well and was much copied, also outside the field of neutron scattering. When he published it in *Journal of Applied Crystallography* **2** (1969) 65, it was however under his name alone. For that reason it became known as The Rietveld Method. [A book with this title, edited by R.A. Young and counting 308 pages, was published in Oxford in 1993]. The contribution of his colleagues was thereby destined to the dustbin of history.

By analyzing the diffraction peaks it had become clear that the always present diffuse background was due to irregularity in the crystals, which becomes strong when the crystals are melting and become liquid. The diffuse background in the scattering by liquids was already under investigation in Delft, at the Higher Education Reactor, where the fluxes were an order smaller than in Petten. It made little sense to compete with the Delft people and copy their work. After completing the mentioned thesis, Cor van Dijk started a project with Wim van der Lugt and Sieb Radelaar in Groningen, who wanted to study the clustering of atoms in liquid metals and metallic alloys. This collaboration included a thesis by Jan Vrijen on clustering in a copper–nickel alloy, resulting from hour–long heating at 400°C, as well as ordering effects in molten alkali–metals and their alloys.

It was not only in elastic neutron scattering that the Petten people were competing with those in Delft, it was also in inelastic scattering. But only in Petten the neutron flux was sufficiently high for detailed studies of the orderly lattice dynamics of solids. So a triple–axis crystal spectrometer was built and installed in beam 1. Between 1965, when this showpiece was completed, and 1970, when the first PhD theses based on measurements with that instrument could be defended, Jitze and Cor van Dijk studied phonon dispersions in magnesium–tin, zinkblende,  $\alpha$ –iron and an iron–aluminium alloy. [We mentioned them already.] Having shown that lattices are moving in an orderly fashion, linked to the crystal structure, also shown in Chalk River, Harwell, Risø and other institutes with a large reactor, the question came up how phonons would propagate close to, or in, a phase transition. They wouldn't fit to the lattice any more. In 1978 Cees de Pater could discuss the situation of non–fitting [incommensurate] worlds, such as present in the phase transitions of Rb<sub>2</sub>ZnBr<sub>4</sub> and Na<sub>2</sub>CO<sub>3</sub>.

All these new findings showed some of the advantages neutron scattering had over X–ray scattering [the latter being affected by the extent of electron clouds around the pointlike nuclei], but the true advantage lay in the fact that nuclei have a spin, and would thus also interact with neutron spins if one were able to align them in a magnetic field, i.e. to polarize them. Adequate apparatus was built in beam 3. In 1964 it consisted of a

single crystal of 92% cobalt and 8% iron, which also selected the neutron momentum. The diffraction of these neutrons is very sensitive for the spin direction and can give information on spin densities as a function of the crystal structure. In 1968 Bob van Laar got his PhD for the determination of simple magnetic structures, and continued this type of work in the *seventies* and also in the *eighties* to unravel complicated structures, often in collaboration with colleagues from Poland.

How useful this work may have been for the advancement of crystallography, it couldn't tell anything about spin behaviour. There was the unexpected! Didn't we start this chapter with the story of spin flipping? For the study of spin dynamics a second instrument was put in beam 3, first a time-of-flight spectrometer, and then a triple-axis spectrometer, with which energy transfers in the scattering could be measured, thus also motions. Evert Frikkee was soon able to publish that he had seen spin waves [*Physica* **32** (1966) 2149]. In 1973 this type of inelastic scattering led to his PhD thesis on spin dynamics in ferromagnetic nickel, not without an advanced theoretical explanation. The next years he co-authored seven papers in *Physical Review* on dynamical phenomena, in which unknown states and transitions in various magnetic substances were brought to light. The confirmation of Wilson's renormalisation theory was only a by-product.

\* Translation of part of a chapter with this title in *De republiek der kernegeleerden*, BetaText, Bergen NH (2000) ISBN 90 75541 01 5.