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National facility for neutron beam research

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Abstract. In this talk, the growth of neutron beam research (NBR) in India over the past five decades is traced beginning with research at Apsara. A range of problems in condensed matter physics could be studied at CIRUS, followed by sophisticated indegenous instrumentation and research at Dhruva. The talk ends with an overview of current scenario of NBR world-wide and future of Indian activities.

Keywords. Reactors; neutron scattering; neutron diffraction; inelastic neutron scattering; neutron spectrometers.

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1. Introduction

Bhabha Atomic Research Centre (BARC) at Trombay is the only place in India where high flux thermal research reactors have been established since the inception of Department of Atomic Energy (DAE). These research reactors find their utilisation in isotope production, engineering research, activation analysis and for providing thermal neutron beams for use in physics research by neutron scattering.

What is so important about neutron scattering? The high penetrating power of neutrons, coupled with their unique sensing capabilities of light atoms among heavier atoms, their wavelength comparable to that of X-rays, capability of exchanging energies in almost similar magnitudes as their normal energies, their magnetic moments making them magnetic sensors and their widely varying cross-sections as one goes from one isotope to another across the periodic table, have made them unique probes amongst all the available probes. The thermal neutron beams have therefore been used extensively over the past five decades for condensed matter research. To determine and understand the structure and dynamics of constituents (electrons/atoms/molecules) of materials there is only one way, namely to study the scattering of radiation from the sample. Diffraction leads to structure determination and inelastic scattering leads to understanding of dynamics of constituents of materials.

What are the resources available to researchers in India for use of neutrons? It is nearly 45 years since study of solids and liquids began with meager facilities at the then Atomic Energy Establishment Trombay (renamed Bhabha Atomic Research Centre or BARC), using India's first nuclear reactor, Apsara. Over the following decades, CIRUS and Dhruva reactors became operational at BARC. Progressively, their design took into account the needs of the neutron scattering community also. These research reactors are being used not only by scientists of BARC but also by a larger number of researchers in the colleges, universities and other institutions of higher learning.

The research facilities especially at Dhruva got into commissioning over the past two decades under a number of five-year plan projects. When we were in the first of these plan periods, some of us wondered whether we can import spectrometers or the neutron guides as is done in many overseas laboratories and mooted the idea. I remember what P K Iyengar, our group leader, said in one of our meetings. He said "well, if you think that you would like to do neutron scattering research by importing the instruments, better you forget that kind of research!" The only way was to get, no doubt, some components or other things but the instruments had to be designed and developed indigenously. From small beginnings, learning all along the way, we graduated from wax-and-string type of instruments to fully engineered instruments.

2. The Apsara days

Apsara reactor (a low flux, enriched U, light water moderated, light water cooled reactor, max rated thermal power 1 MW, max central thermal neutron flux $\sim 10^{12}$ neutrons/cm^2/s), commissioned in 1956, operating at around 300 kW provides neutron beams for neutron beam research (NBR). The special facilities at this reactor include the shielding corner, a thermal column and irradiation space within and around the core. My mind goes back to 1958, when I was a beginner in this field. Specifically related to NBR, a home-built single crystal neutron spectrometer was set up at Apsara around 1956. Total cross-sections of a few elemental solids were measured using this spectrometer over a very narrow energy range. This instrument was also used for studying the thermal neutron spectrum of neutrons as diffracted from a variety of large mosaic single crystals. It turned out that these measurements led to the observation of neutron double Bragg scattering for the first time: the monochromatic spectrum from a single crystal was not a smoothly varying function of neutron wavelength but instead there were deep valleys and peaks in the measured spectrum. I was involved in these first observations. The experiments had to be carried out, literally manually, by using the simple single crystal spectrometer, a lamp, mirror-and-scale arrangement for effecting angular changes, a stopwatch and a scaler counter. The experiments taught me, if nothing else, patience and perseverance; it was exciting to me because I was measuring something that others had not observed with neutrons so far. Although the observations were wrongly attributed to inelastic scattering effects to begin with, it was soon realised that the effect was analogous to 'Renniger effect' well-known in X-ray diffraction. The other experiments carried out using this instrument were the study of the shape of thermal neutron resonances in metals like In. The broad resonances could be explained on the basis of Debye spectrum of phonons in the material. Apsara is still in use even after nearly forty-five years, now for neutron radiography, for testing detectors and for bulk shielding experiments connected with shield design of prototype fast breeder reactor.

By late fifties, a diffractometer was imported from England. This instrument was a copy of the instrument used by G E Bacon at Atomic Energy Research Establishment, Harwell and was referred to as a 'Curran' instrument, 'named after John Curran of Cardiff whose main business was accurate manufacture of radar turntables so they knew he would be able to make them a good goniometer'. When Iyengar returned from Chalk River Nuclear Laboratories (*née*, Atomic Energy of Canada Ltd.), he converted this instrument rather ingeniously to carry out phonon measurements. In addition, a diffractometer was developed based on a gun-mount obtained from the naval docks; this was used for the first unpolarised magnetic diffraction studies on a Fe–Ge alloy.

One notices that even today, experiments and researches start at rudimentary levels at many a place. So one need not shy away from such beginnings. Our instruments have metamorphosed to such an extent that when we compare today's instruments at Dhruva with those at Apsara or later on at CIRUS reactors, we may not find much resemblance to their ancestors!

3. The CIRUS days

When CIRUS (a medium flux, natural U, heavy water moderated, light water cooled reactor; max rated thermal power 40 MW, max central thermal neutron flux $\sim 6 \times 10^{13}$ neutrons/cm^2/s) got commissioned in 1960, trained manpower was available for effective utilisation of this reactor, to initiate large-scale programmes in neutron crystallography, magnetic diffraction, phonon and magnon spectroscopy and study of dynamics in liquids and molecular systems. Under the leadership of Iyengar, a large number and variety of neutron spectrometers were designed and built at Trombay. These were provided with data acquisition systems that could gather data automatically round the clock using electromechanical control systems. The first fully automated computer controlled diffractometer was designed and commissioned by R Chidambaram and coworkers.

Around 1965, several spectrometers were functional at CIRUS. Techniques like triple-axis spectrometry, time-of-flight spectrometry, polarised neutron diffraction etc. had become integral to NBR. The multi-arm spectrometer was a unique instrument anywhere in the world. The entire experimental facilities were home-built.

CIRUS reactor hall was a beehive of activity with some 20 researchers along with the supporting staff. A variety of new experimental investigations on par with international investigations were realised soon. Some of the measurements were carried out using indigenous cryostats using liquid nitrogen. For specific magnetic studies liquid helium transported from TIFR was also used in a helium cryostat specially designed and built for neutron investigations. N S Satya Murthy led a group specialised in the use of polarised neutrons. They built a polarised neutron diffractometer, which provided unique results in several ferrites, heusler alloys and other dilute magnetic systems. G Venkataraman and K Usha Deniz designed and developed the rotating crystal time-of-flight spectrometer with its own time-of-flight 'kick-sorter'. This instrument helped in studying quasi-elastic and inelastic scattering from molecular systems using pulsed monochromatic cold neutrons. C L Thaper was operating a Be-filtered inelastic neutron spectrometer. B A Dasannacharya, P S Goyal and others developed the first small angle neutron spectrometer based on Be-filtered neutrons at CIRUS, in addition to pursuing other research problems.

Some of the interesting findings of the CIRUS days are the following:

- Determination of crystal structure of amino acids.
- Study of nature of hydrogen bonding in hydrogenous crystals, flexibility of hydrogen bonds and their relevance to biological systems.
- Measurement of phonon dispersion relations in Be, Mg and Zn.
- Investigations of Kohn anomalies in Zn.
- Molecular reorientation studies in NH_4 halides by TOF technique and by filter detector spectrometry.
- Diffraction from liquids.
- Development of high resolution inelastic scattering techniques like difference filter technique.
- Polarised neutron diffraction from magnetic materials.
- Magnetisation density, magnetic form factors and spin wave dispersions in single crystals.
- First observation of non-collinear Yafet–Kittel-type of ordering in a mixed ferrite.
- Study of anharmonicity in librational potential in molecular systems.
- First measurement of phonons in an ionic 'molecular' crystal α-KNO_3 and so on.

The CIRUS days also saw our laboratory collaborating with some countries in Asia. The Regional Cooperation Agreement (RCA) concept, under IAEA, originated as a result of these activities.

4. On with Dhruva

Substantial activities from CIRUS got shifted after 1985 to Dhruva reactor (a medium flux, natural U, heavy water moderated, heavy water cooled reactor; max thermal power 100 MW, max central thermal neutron flux $\sim 2 \times 10^{14}$ neutrons/cm^2/s), an indigenously built reactor. Dhruva is the main workhorse for neutron scattering programme since then. The reactor has tailored features for NBR like recessed cavities for installing neutron spectrometers close to the core, cavities for installing neutron monochromators and neutron guides in the biological shield, tangential beam tubes and large diameter beam tubes for installing cold and hot moderators. In addition, a large adjoining hall was built to take cold neutron beams from the reactor to this hall through neutron guides.

With the availability of Dhruva reactor for NBR, engineering design of a prototype triple-axis neutron spectrometer was achieved. This model spectrometer, shown in figure 1, could be suitably modified to cater to needs of various subdisciplines of NBR. P R Vijayaraghavan played a crucial role in building and commissioning the spectrometers at the reactor with help from Nuclear Physics Division workshop, Central Workshops, Reactor and Reactor Operations & Maintenance

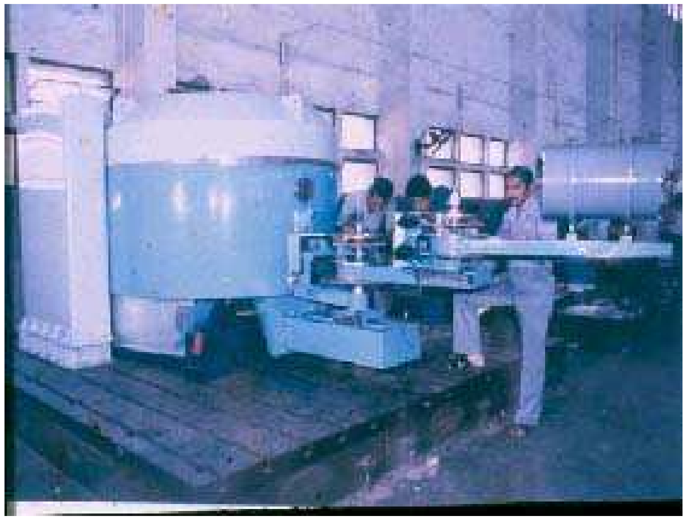

Figure 1. Prototype triple-axis spectrometer under test.

Groups. As far as technology improvement was concerned, apart from design and manufacture of the spectrometers based on the proto-type one mentioned above, one might note that important innovations and inputs related to full automation of control and data acquisition system on each spectrometer using personal computers. It was no mean achievement that a computer controlled triple-axis spectrometer was fabricated and exported to Bangladesh under a purchase order from IAEA. Vijayaraghavan was responsible for installing and commissioning this instrument to the satisfaction of scientists in Bangladesh in a matter of three weeks. This instrument could be used for powder diffraction, inelastic spectroscopy and for small angle scattering by suitable but minor modifications.

Another major activity related to design, development, fabrication and installation of neutron guides at this reactor by L Madhav Rao with support from TPPED and other agencies. Starting from scratch, namely, from import of float glass plates, one had to get various hardware inputs like fine nickel wire, tungsten filaments, design and development of a stainless steel framework as a holding device etc., to enable nickel coating of some 400 glass plates. A high vacuum coating plant was available, kind courtesy of the Indian Institute of Astrophysics at Kavalur. Assembly of guides each 1 m long and then mounting and aligning these over nearly 30 m to high precision and finally enclosing these guides with adequate shielding were the tasks quite painstakingly carried out totally in-house by Madhav Rao and Shukla. Figure 2 shows a picture of guides under installation. By 1990, the layout of instruments at Dhruva was somewhat like what is shown in figure 3.

We shall end this historical note by mentioning the salient physics that was achieved during the eighties and nineties without going into any detail:

- Determination of crystal structure of nearly a hundred varieties of high temperature superconductors (A Sequiera and coworkers).

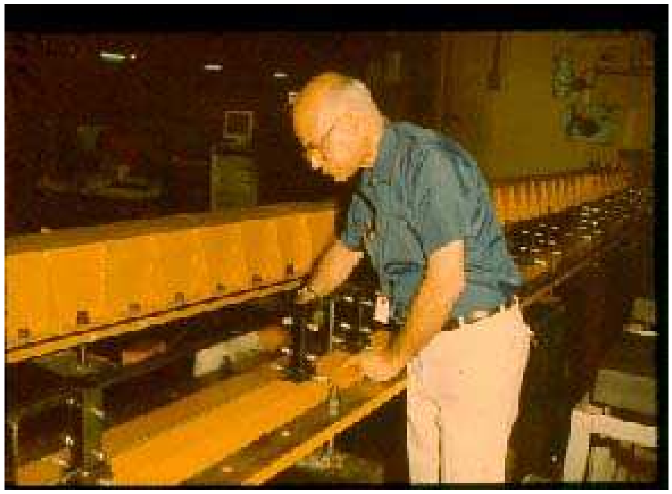

Figure 2. Neutron guides under installation at Dhruva reactor.

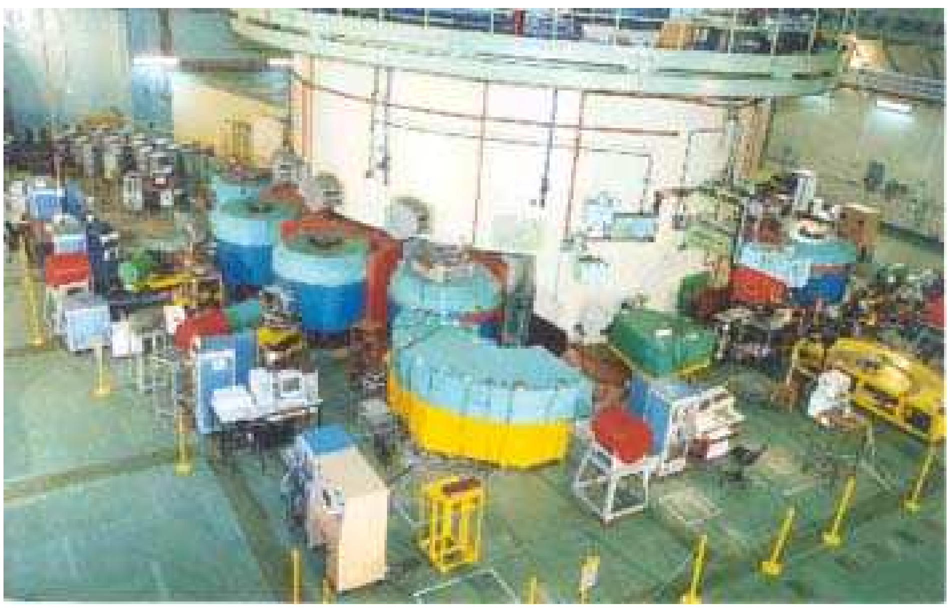

Figure 3. Layout of spectrometers at Dhruva reactor hall.

- Determination of magnetic structure of a number of systems in the range of temperature of 10–300 K (S K Paranjpe and coworkers).
- Small angle scattering studies of micelles and surfactants (B A Dasannacharya and coworkers using a SANS at CIRUS).
- Inelastic scattering studies of phonons in minerals, magnetic materials, high T_c superconductors etc. (S L Chaplot and coworkers).

- Study of reorientational motions in various 'molecular' systems by high-resolution quasi-elastic neutron scattering (R Mukhopadhyay and coworkers).
- Polarisation analysis and other magnetic studies (L Madhav Rao and coworkers).
- Study of disordered systems (P S R Krishna and coworkers).

The experimental results were quite different from those that were merely the logical extensions of the work carried out at CIRUS. In addition, there were some important theoretical investigations also. During the past six to eight years, additional spectrometers have been installed in the Dhruva guide hall. A variety of new investigations have fructified. Some recent studies from Dhruva NBR are given below:

(i) Powder diffraction studies have related to:

- Ferrites having magnetic disorder
- Manganites showing colossal magnetic resistance
- Cobaltates showing ferro and antiferromagnetic behaviour
- Titanates and zirconates exhibiting ferro/antiferroelectric behaviour
- Alloys of transition metals, rare earths and actinides showing exotic magnetic properties.

(ii) Quasi-elastic scattering studies using MARX (multi angle reflecting crystal) spectrometer have related to:

- Molecular dynamics of different organic molecules in zeolite-like cage structures
- Molecular reorientations of alkyl chains in monolayer protected clusters
- Molecular reorientations in organic and inorganic salts
- Molecular reorientations in liquid crystals, microemulsions, etc.

(iii) Inelastic spectroscopy:

- Study of complex materials, especially of minerals, has continued based on a combination of inelastic scattering experiments, calculations of phonon spectra and computer simulation experiments
- A noteworthy experiment in collaboration with ILL scientists related to *negative thermal expansion (NTE)* of cubic zirconium tungstate: High pressure inelastic scattering measurements on IN6 up to 1.7 kbar showed a pronounced softening of the phonon spectrum below 8 meV by $\sim$0.1 to 0.2 meV. This unusual softening accounts for observed NTE over the entire range of temperature.

(iv) Studies carried out using polarisation analysis spectrometer:

- U-, Th-, and Ce-based intermetallic compounds
- Disordered mixed ferrites
- Amorphous magnetic materials

- Geometrically frustrated compounds
- Colossal magnetoresistant perovskites
- Molecular magnetism.

(v) SANS from large inhomogeneities using double crystal-based small angle spectrometer:

- Pore morphology and pore-matrix interface roughening of metamorphosed sedimentary rocks, sandstones and igneous rock samples
- Ceramic sintering

(vi) Polarised neutron reflectometry:

Polarised neutron reflectometry (PNR) is an important non-destructive tool to determine chemical as well as magnetic structure of thin film samples. Using this technique the following studies have taken place:

- reflectivity profile from $[(\mathrm{Ni–Mo\ alloy})/\mathrm{Ti}]_{10}$ on glass substrate
- determination of Fe moment in a Fe/Ge multilayer sample

(vii) In neutron interferometry and optics, V C Rakhecha and A G Wagh made notable contributions. They brought to bear rare and new insights, which in some instances questioned rightly the interpretations of experimental results by western experts; the results were published in a series of papers in Letters sections of various journals. Their work spanned esoteric phenomena like Pauli anticommutation, observation of non-cyclic phases and separation of geometric and dynamical phases. In addition to theoretical interpretations, they carried out experiments on neutron interferometry in a few overseas laboratories. Much of this work is beyond my limited comprehension. But I recently noted a piece of their work related to 'producing a monochromatic beam with the sharpest angular profile', which I found interesting. I shall quote from their paper in IANCAS Bulletin: '... Our results [A G Wagh, V C Rakhecha and W Treimer, *Phys. Rev. Lett.* **87**, 125504 (2001); *Appl. Phys. A* (2002)] mark well over an order of magnitude improvement over all Bonse–Hart multiple Bragg reflection profiles reported previously. We have thus produced a monochromatic neutron beam with the sharpest angular profile in the world. This work opens new possibilities for SUSANS (super ultra-small angle neutron scattering) studies enabling characterising of samples containing micron-size agglomerates'.

5. Collaborative programmes and the outlook

When the new spectrometers got in place and research activities gathered momentum at Dhruva, two important developments occurred: (a) When it was suggested that some plan money may be made available to our scientists for traveling overseas labs for carrying out advanced experiments, mostly based on preliminary studies carried out at Dhruva, Chidambaram and Iyengar readily supported the idea. Many eyebrows were raised, because this was not something done! To think of visiting another laboratory for one or two days' beam time appeared strange. But we could get the project approved. Then onwards this avenue has been used by many in Solid State Physics Division to visit many labs for carrying out experiments for short durations. The benefits were that our scientists could literally lay hands on advanced instruments at the higher flux neutron facilities and gain knowledge and information on the state-of-the-art instrumentation and techniques and also carry out research on materials of our interest. In a way, this kept our colleagues out of obsolescence. Needless for me to point out that this approach was a cost-effective impetus to on-going research programmes. We could collaborate very effectively, especially with ISIS at Rutherford Appleton Lab, UK, by providing a Day-1 instrument, namely, a ΔT spectrometer and later on with OSIRIS also. This was a unique way of paying by kind instead of by cash for utilization of the spallation source at ISIS. It must be underscored that many a lab in several European countries or labs in Japan and other countries have been contributing in cash, enlisting as members, for utilisation of several neutron and synchrotron facilities all over Europe and at other places. Perhaps India is the only country that has benefited much from this part of the world in such international endeavours. (b) Another important development of the late eighties was the establishment of collaborative programmes with the students and faculty in our universities. DAE opened the doors of its major research facilities – Dhruva at BARC, Variable Energy Cyclotron at Kolkata and the INDUS- I synchrotron at Indore – to the university community by setting up the Inter-University Consortium for DAE Facilities (IUC-DAEF) in collaboration with the University Grants Commission. Under this joint programme, one of the IUC-DAEF centers was set up at BARC. The programme involved training of research students in NBR and supporting joint research programmes. Nearly 30 projects were running at any one time and Dhruva became truly a national facility. This programme has been mutually beneficial for furthering research in various materials by NBR in our country. Whereas synthesis of new and novel materials, their characterisation and non-neutronic studies were undertaken in the universities, their study by NBR was undertaken at BARC and the interpretation and understanding of the data was a cooperative joint endeavour. It has given scope to bring in young researchers, new materials, fresh ideas and NBR expertise together for important studies. To me, success of Dhruva as a national facility has crucially depended on the success of IUC-DAEF as an equal-partner programme. It is gratifying to learn at this meeting from Anil Kakodkar that a fresh MOU is signed between DAE and UGC to extend and enhance the consortium activities.

As far as the current international scenario is concerned, one may note that the most important project is the ongoing construction of $1.4 billion Spallation Neutron Source (SNS) at Oakridge, a single-target, 2-megawatt facility that is expected to be operational in 2006. Multi-million dollar upgrades have taken place at: Los Alamos Neutron Science Center (LANSCE), Intense Pulsed Neutron Source (IPNS) at Argonne and at High Flux Isotope Reactor (HFIR) at Oakridge. Intense activity in building or designing next-generation neutron sources in European Union, Japan (a new 1-MW spallation source is expected to come online in 2007), China and Australia are in place.

As far as the outlook for the future is concerned, 'as we enter the new millennium, we face the 'Age of Designer Materials', designed molecule by molecule, either on the computer or in the laboratory involving integration of synthesis and simulation of complex materials'. "Whoever controls the materials, controls the science and the technology" stated J Wade Gilley, President of The University of Tennessee. Neutron scattering will continue to help understand as new materials are ushered in. Materials research will continue unabated.

Another aspect that I like to point out is that neutron scattering techniques are not fully exploited even after 50 years since the beginnings of NBR. (a) In 1963, Martin Blume in US and Malyev *et al* in the USSR discussed general expressions for polarisation dependent inelastic cross-sections. As of 2001, only two experiments had been performed: (i) by Regnault *et al*, *Physica* (1999) and (ii) by Caciuffo *et al* (2001). (b) Total cross-sections up to, say, 70 A^{-1} range are measurable on only very few instruments. Hence there is scope to exploit such measurements as pointed out recently by Martin Dove, *Euro J. Mineral* **14**, 331–348 (2002). (c) Roger Cowley has drawn attention very recently [*J. Phys.* **15**, 4143 (2003)] as to how little we know of the nature of epithermal neutron spectrum at spallation neutron sources and limitation of our understanding of various corrections that need to be made in data analyses. Hence quantitative and correct cross-sections are still not available. These are a few examples that show immense possibilities for neutron to investigate new materials and new phenomena. Just as X-rays are flourishing beyond 100 years after their discovery, neutrons will continue their glorious innings.

Acknowledgements

I am grateful to many of my erstwhile colleagues for sending me information liberally to prepare this talk.