ICANS-XV
15th Meeting of the International Collaboration on Advanced Neutron Sources
November 6-9, 2000
Tsukuba, Japan

3.4
Neutron beam research at BARC (India) and international collaboration

M. Ramanadham* and R. Mukhopadhyay**

Solid State Physics Division, Bhabha Atomic Research Centre, Mumbai 400085, India
E-mail: ramu@magnum.barc.ernet.in, **E-mail: mukhop@apsara.barc.ernet.in

Abstract

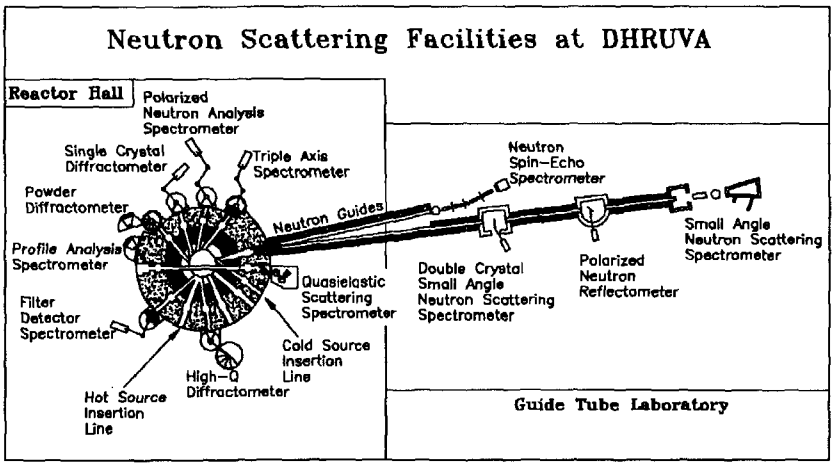
Neutron beam research started in India more than four decades ago. Presently, the National Facility for Neutron Beam Research, NFNBR is located in *Dhruva*, a 100 MW research reactor. The entire facility, including the development of neutron detectors, is the result of indigenous efforts of the participating scientists from Bhabha Atomic Research Centre, BARC. NFNBR is accessible to national and international collaborations, and about forty research groups from various institutions have already availed this facility. Active collaboration with ISIS started since 1984, when the day-1 spectrometer, built at BARC, became operational at ISIS. The collaboration continued with the fabrication, at BARC, of parts for OSIRIS spectrometer. Many neutron beam researchers from BARC have carried out collaborative experiments using the neutron sources at USA, France, Germany, Switzerland, and Japan.

Introduction

Neutron scattering at Trombay started with the facility at APSARA reactor (100kW) in 1956, and then continued with CIRUS (40 MW), which became available in 1960 and then the present reactor DHRUVA. Dhruva is a 100 MW natural uranium reactor with peak thermal neutron flux of 1.8×10^{14} neutrons cm^{-2} s^{-1}, tailor-made for neutron scattering experiments with tangential beam holes, through-tube, separate moderators for cold and hot neutrons, guide-tube, etc. Various spectrometers, like triple-axis, polarization analysis, single-crystal and powder diffractometers, Hi-Q diffractometer, QENS spectrometer, SANS instruments, spin-echo spectrometer, reflectometer, etc., are installed. This facility is open for national and international collaborations. All the instruments have been fabricated in the in-house workshops, and are controlled by personal computers. State of the art technology was used to design and fabricate the spectrometers. 1-D and 2- D position sensitive detectors (PSD), and associated electronics have also been developed indigenously. Many of the spectrometers are with PSDs, providing higher throughput required in a medium flux reactor. One of the spectrometers (QENS spectrometer) is installed on 'Tanzboden' (air-cushion), also developed in-house, facilitating easy maneuverability, which also has vertically curved monochromator for focusing neutrons. Other than our own National Facility for Neutron Beam Research, NFNBR, we have collaboration with ISIS facility at Rutherford Appleton Laboratory, UK since 1984. Our perspective has been to carry out experiments at BARC, and then follow-up with the ISIS collaboration for more sophisticated studies. This has worked very well, and resulted in the fruitful BARC-RAL collaboration for last 15 years.

Neutron scattering facilities at BARC

Figure.1 shows the general layout in the reactor hall and the guide-tube laboratory. There are four spectrometers at the tangential beam lines, two at the ends of the through tube, two at the radial beam lines looking at the hot source, and four instruments in the guide laboratory.



The present-day facilities include, single-crystal and powder diffractometers, polarization analysis spectrometer, Hi-Q diffractometer, triple-axes & filter-detector spectrometers, quasi-elastic scattering spectrometer, all installed in the reactor hall, and two small-angle scattering instruments, spin-echo spectrometer and reflectometer (under commissioning) in the guide-tube laboratory. Liquid methane at 100 K and graphite at 1800 K (achieved by nuclear heating) are the cold and hot sources respectively, under development.

Inelastic scattering studies: The inelastic neutron scattering activities essentially involve (i) experiments using the triple-axis spectrometer (TAS) at BARC, and (ii) extensive international collaborations involving INS measurements using steady-state reactor sources at the Brookhaven National Laboratory, Oak Ridge National Laboratory, USA, etc., as well as spallation sources like the ISIS, RAL, etc. The systems studied find several useful applications, such as, the measurements of the phonon dispersion relations and density of states of several geophysically important minerals like forsterite Mg_2SiO_4 [1], fayalite Fe_2SiO_4 [2], enstatite $Mg_2Si_2O_6$ [3], the garnet mineral almandine $Fe_3Al_2Si_3O_{12}$ [4], the aluminosilicate $Al2SiO_5$, minerals sillimanite, andalusite and kyanite [5], and the mineral zircon $ZrSiO_4$ [6], the measurements of the phonon density of states in intermetallics like Zr_2Ni, Zr_2Fe, and the studies of magnetic excitations in $CeSn_2In$, $CeSi_{2-x}Ga_x$ etc. INS measurements, backed by theoretical lattice dynamics calculations have provided microscopic insights into the nature of phonon dispersion relations and density of states in various mineral phases in the Earth's mantle. The calculations have enabled planning, execution and analysis of the experiments and have helped in an atomic level understanding of the experimental data.

Quasi-elastic scattering studies: The QENS spectrometer (in MARX mode, $\Delta E/E = 4\ \%$) is novel in many aspects among the neutron spectrometers at Dhruva [7]; (i) It is the only instrument installed on 'Tanzboden' (air cushion), (ii) vertically bent monochromator is used to focus neutrons, (iii) monochoromators are located inside a cavity of the biological shield. This medium resolution spectrometer ($\Delta E/E \sim 200\mu eV$) is used to study problems suitable for it. The systems studied include, molecular motions in various molecular solids, namely, reorientation of pyridinium ion in Pyridinium halides [8], molecular motions in liquid crystal BBBA (4O.4) [9], Ammonium ion reorientations in $(NH_4)_2SbF_5$ [10], dynamics of water in

porous medium like alumina gel [11] or silica gel, lignite coal [12], cement etc., and dynamics of hydrocarbon in confined geometry like dynamics of Propane in Na-Y Zeolite, and benzene in HZSM-Zeolite.

Single-crystal diffraction: All along, the research activities of single-crystal neutron diffractometry were centered around the studies on hydrogen-bonded crystal structures. Earlier experiments were on a number of inorganic hydrates, which rendered possible a detailed analysis of the hydrogen bonding and lone-pair coordination around the water molecule [13]. One of the most significant achievements during the late sixties and early seventies was the determination of crystal structures directly from neutron diffraction data using the direct methods of solving the phase problem, in spite of the presence of negative scatters in the crystal structure [14,15]. Structural studies on amino acids and small peptides were taken up since 1969, and a comprehensive H-bond analysis in bio-molecules was carried out [16], using these and similar studies reported from BNL, USA and elsewhere. Results, thus obtained, were used very effectively, in the interpretation of x-ray protein structures [17] and ferroelectric phase transitions [18]. Present studies include those on protein and DNA complexes, ferroelectrics and optoelectronic materials.

Powder diffraction: The profile analysis spectrometer ($\delta d/d$ = 1.3%) has been used extensively for the study of structural and magnetic phase transitions in a variety of oxides, alloys and intermetallic compounds. Observations of unconventional ferrimagnetism in disordered spinel $Zn_xCo_{1-x}FeCrO_4$ ($0.45 \leq x \leq 0.55$) [19], transformation from metallic ferromagnet to insulating antiferromagnet in $La_{0.7}Sr_{0.3}Co_{1-y}Fe_yO_3$ ($0 \leq y \leq 1$) with increase in Fe content [20], superlattice reflection in $Sr_{1-x}Ca_xTiO_3$ ($x \geq 0.12$) and $Pb_{1-x}Ba_xZrO_3$ (x = 0.2 and 0.3) [21], non-collinear magnetic ordering in $ErFe_2H_x$ justifying the conjecture of *fanning* of moments [22], long range ferromagnetic order in $UFeMnSi_2$, collinear ferromagnetic ordering in orthorhombic $UNiSi_2$, coexistence of antiferromagnetic and spin glass order at low temperatures in $Cr_{60-x}Fe_{20}Mn_x$ [23] are some typical results.

Magnetic studies using polarized neutrons: A neutron polarization analysis spectrometer (PAS) is used for magnetic scattering studies [24]. This spectrometer can be used in different modes, such as (a) diffraction mode (2-axis configuration), (b) polarization analysis mode (3–axis configuration in scattering geometry), and depolarization mode (3-axis configuration in transmission geometry). It has been used on various magnetic systems such as anisotropic $U_{1-x}Th_xCu_2Ge_2$ intermetallic compounds [25], superparamagnetic Ce_2Fe_{17} alloy [26], disordered ferrites $Zn_{1-x}Co_xFe_{2-y}Cr_yO_4$ ($x \sim 0.5$ and $y \sim 0.9$) [27], geometrically frustrated $KMnFeF_6$ system [28], amorphous re-entrant spin glass $Fe_{90-x}Ru_xZr_{10}$ (x = 0, 1 and 5) alloys [29], colossal magnetoresistance (CMR) materials such as, $La_{0.67}Ca_{0.3}Mn_{0.9}Fe_{0.1}O_3$ [30] in order to understand the magnetic nature of magnetic ordering in these systems.

Hi-Q diffractometer: Hi-Q diffractometer is a multi-PSD based instrument covering Q-range up to 14 $Å^{-1}$. It is used for structural studies in amorphous and liquid systems. Typical recent studies include, structural and Network connectivity in Ge_xSe_{1-x} glasses [31], hydrogen bonded molecular clusters in liquid CD_3OD [32] and diffraction studies of Rare Earth Phosphate glasses [33] etc.

Small-angle scattering: A SANS diffractometer at the guide-tube laboratory has been installed at the end of the guide G1 (Fig.1). This guide has a cut-off wavelength, λ^* = 2.2 Å. The neutron beam from the guide is monochromatized by the BeO filter. The average

wavelength of the monochromated beam is 5.2 Å and has a spread ($\Delta\lambda/\lambda$) of about 15 %. The Q range of the diffractometer is 0.018 - 0.30 $Å^{-1}$ and it is suitable for inhomogeneities of the sizes in the range 10 – 150 Å. The structural aspects of a variety of micellar solutions have been studied using SANS at BARC, which includes conventional surfactants [34-36], mixed surfactants [37], gemini surfactants [38], block copolymers [39] and surfactants with multiple head groups [40].

There is another double crystal based moderate resolution small-angle neutron scattering instrument has been built and commissioned at the guide tube laboratory. The instrument consists of a non-dispersive (1, -1) setting of (111) reflections of silicon single crystals with sample between the two crystals. The used neutron wavelength is 0.312 nm. The analyzer crystal rotates with smallest step size of 0.0012°. The accessible range of wave vector transfer q is 0.003- 0.173 nm^{-1}. This type of facility has found fruitful applications in ceramics and cements [41].

International Collaboration

BARC has been collaborating with ISIS facility, Rutherford Appleton Laboratory, UK since early Eighties. A spectrometer referred as 'ΔT window' spectrometer was designed, fabricated and supplied to ISIS had been the day-1 spectrometer in 1984 [42]. The 'temperature difference window' spectrometer was a high-resolution inelastic spectrometer. This was later replaced by a high efficiency crystal analyzer spectrometer, IRIS being used at present. The collaboration has been renewed with our participation in the OSIRIS spectrometer. It is a full polarization analysis inelastic spectrometer cum diffractometer. Part components of this spectrometer are fabricated in BARC. OSIRIS is funded by Spain, Sweden, Switzerland, Italy, UK and other participating countries.

We have been a regular user of the ISIS facility for carrying out neutron experiments for last 15 years. This resulted to more than 50 publications in international journals [43]. At present we have active collaboration with, 1) SINQ, Switzerland, 2) HMI, Berlin, 3) ANL, USA, 4) ILL, France, 5) KEK, Japan towards various scientific problems which includes, Interferometry, SANS, INS, QENS etc It has been our perspective to carry out experiments with our own facility and follow-up with international facilities by submitting proposal through peer review.
BARC have also collaborated with many third world countries since early sixties. Neutron scattering instruments (designed and fabricated at BARC) were supplied to Philippines, South Korea and Bangladesh. Scientists from various labs of southeast countries were trained by exchange of visits of scientists and active scientific collaborations.

Acknowledgements

We gratefully acknowledge the numerous contributions from colleagues, past and present, to the neutron beam research in India. We thank Dr. S.K. Sikka for his support, encouragement and interest in our endeavor.

References

[1] K.R. Rao *et al*, Science, **236** (1987) 64; Phys. Chem. Min. **16** (1988) 83.
[2] S. Ghose *et al*, Physica B, **174** (1991) 83.

[3] N. Choudhury N. *et al*, Phys. Rev., **B58** (1998) 756.
[4] R. Mittal *et al.*, Phys Rev., **B61** (2000) 3983.
[5] Mala N. Rao *et al*, Phys. Rev., **B 60** (1999) 12061.
[6] R. Mittal *et al.* Phys. Rev., **B 62** (2000) In press.
[7] R. Mukhopadhyay and S. Mitra Nucl. Inst. Meth. A (2000) In press.
[8] S. Mitra *et al*, Sol. State Phys. (India) 2000.
[9] S. Mitra, R. Mukhopadhyay and K. Venu, Chem. Phys., **261** (2000) 149.
[10] R. Mukhopadhyay, P.S. Goyal and C.J. Carlile, Phys. Rev. B, **48** (1993) 2880.
[11] S. Mitra *et al*, Solid State Commun., **105** (1998) 719; J. Non. Cryst. Solids, **235-237** (1998) 229.
[12] S. Mitra, R. Mukhopadhyay and K.S. Chandrasekaran Physica B, **292** (2000) 29.
[13] R. Chidambaram, A. Sequeira & S.K. Sikka, J. Chem. Phys. **41** (1964) 3616.
[14] S.K. Sikka, Acta Cryst., **A25** (1969) 539.
[15] S.K. Sikka, Acta Cryst., **A26** (1970) 662.
[16] M. Ramanadham & R. Chidambaram, in: Advances in Crystallography, (Ed: R. Srinivasan), Oxford & IBH Publishing Co., New Delhi, India (1978) 81.
[17] M. Ramanadham, V.S. Jakkal & R. Chidambaram, FEBS Lett., **323** (1993) 203.
[18] R. Ranjan-Choudhury, R. Chitra & M. Ramanadham, in: Abstracts of XXX National Seminar on Crystallography, June 28-30, 2000, Tirupati, India.
[19] R. Chakravarthy *et al.*, Phys. Rev. B, **43** (1991) 6031.
[20] V.G. Sathe *et al*, J. Phys.Condens. Matter, **10** (1998) 4045.
[21] Rajeev Ranjan *et al*, J. Phys.: Condens. Matter, **11** (1999) 2233; Appl. Phys. Lett., **74** (1999) 1.
[22] K. Shashikala *et al.*, Phil. Mag. **B79** (1999) 1195.
[23] A. Das, *et al.*, J. Phys.: Condens. Matter, **11** (1999) 5209.
[24] S. M. Yusuf and L. Madhav Rao, Neutron News **8** (1997) 12.
[25] S. M. Yusuf, L. Madhav Rao and P. Raj, Phys. Rev. B, **53** (1996) 28; Solid State Commun. **112** (1999) 207.
[26] S. M. Yusuf, Powder Diff. **8** (1993) 236.
[27] S. M. Yusuf *et al*, J. Phys.: Condens. Matter, 7 (1995) 873; 7 (1995) 5891.
[28] S. M. Yusuf, *et al*, Solid State Commun. **101** (1997) 145.
[29] S. M. Yusuf *et al*, J. Magn. Magn. Mater. **166** (1997) 349.
[30] S. M. Yusuf, *et al*, Phys. Rev. B, **62** (2000) 1118.
[31] N. Ramesh *et al.*, J. Non-cryst. Sol. **240** (1998) 221.
[32] R.N. Joarder *et al.*, Phys. Lett. **253** (1999) 207.
[33] A. Shikerkar *et al*., Journal of Non-Crystalline Solids **270** (2000) 234.
[34] P. S. Goyal *et al*, Chem. Phys. Lett. **211** (1993) 559.
[35] P.S. Goyal, *et al*, Phys. Rev. E, **51** (1995) 2308.
[36] V. K. Aswal *et al*, J. Phys. Chem. B, **102** (1998) 2469; Phys. Rev. E, **61** (2000) 2947.
[37] S. De *et al*, J. Phys. Chem. B, **101** (1997) 5639.
[38] S. De *et al*, J. Phys. Chem. **100** (1996) 11664.
[39] V.K. Aswal *et al*, Phys. Rev. E, 59 (1999) 3116.
[40] N. Jain *et al*, J. Phys. Chem. B, **102** (1998) 8452.
[41] S. Mazumder and A. Sequeira, Pramana - J. Phys. **38** (1992) 95; Phys. Rev. B, 39 (1989) 6370; Phys. Rev. B, **41** (1990) 6272.
[42] P.S. Goyal *et al*, Pramana, **23** (1984) 559.
[43] ISIS Annual reports, 1986-1999.