1. Neutron Scattering Research at JAERI Reactors - Past, Present and Future -

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Abstract

It was in 1961 that the first neutron scattering experiment was performed in Japan at JRR-2. The start of JRR-3 in 1964 accelerated the neutron scattering activities in Japan. The research in this field in Japan grew up by using these two research reactors. Among them JRR-2 has played an important role because its neutron flux was about seven times higher than that of the old JRR-3. The completion of the new JRR-3M in 1990 made an epoch to the neutron scattering activities in Japan. The long-waited JRR-3M came up to the expectations of the scientists of Japan. It is a realization of the ideal reactor with tangential beam holes, cold source and neutron guides in a large guide hall. The flux at the neutron scattering instruments is about five times higher than that of JRR-2. Utilization of JRR-3M has just started. Twelve neutron scattering machines are running there. The number will increase up to close twenty in a couple of years.

INTRODUCTION

The completion of JRR-3M in 1990 made a great epoch to the neutron scattering research in Japan. Great expectation is held by scientists who are interested in neutron scattering experiments.

When the first neutron diffraction experiment was performed at JRR-2 about thirty years preceding the JRR-3M, only two horizontal beam holes were used for neutron scattering facilities. The number of instruments, however, had been increased year by year until when the old JRR-3 was closed in 1983 for the upgrading. Four neutron spectrometers had been installed at JRR-3 and eight instruments had been installed at JRR-2 by that time. Most of them have been repeatedly improved and upgraded to keep up with the very rapid progress of the neutron scattering technology. Out of them, three instruments have been moved to JRR-3M, but six of them are still being used at JRR-2.

The design, construction of those instruments and research activities utilizing them had been done by the great effort of the solid state physicists who were interested in neutron scattering especially in relation with magnetism, ferroelectricity and metallurgy. From the beginning up to now about one third of the beam ports for the neutron scattering have been used

by the Department of Physics of JAERI and the rest two thirds by universities represented by the Institute for Solid State Physics(ISSP) of the University of Tokyo, the Faculty of Science of Tohoku University and the Institute for Materials Research(IMR) of Tohoku University. In this paper, the short history of the neutron scattering activities performed at JAERI reactors, the present status of the neutron scattering facilities there and the future instrumentation plan at JRR-3M are reported.

DAWNING

Neutron scattering experiment in Japan was started as soon as JRR-2 started to operate at low power. That experiment was performed by the solid state physics scientist of JAERI who had been longing for their own thermal neutron beam for a long time. The first experiment done in 1961 was the magnetic structure study of UFe_2 using a polycrystalline sample. It is worth pointing out here that the measurement was made not only at room temperature but also at liquid nitrogen temperature and under applied external magnetic field. The result was reported at the International Conference on Magnetism held in Kyoto in 1961[1]. Two other papers[2,3] were presented there by JAERI and ISSP to report their respective diffractometers built at JRR-2. Therefore the start of the neutron scattering experiment in Japan was about fifteen years after the first substantial neutron diffraction experiment performed at the graphite reactor in Oak Ridge USA.

Prior to this, when it was decided to construct the JRR-2 reactor, a committee was organized in JAERI to study the basic plan of the neutron scattering instrumentation. There it had been decided to construct a double-axis diffractometer in JRR-2. At the almost same time, university scientists started similar committee and it had been decided that ISSP to construct another diffractometer to JRR-2. The Electrical Communication Laboratory of Nippon Telegraph and Telephone Public Corporation also decided to construct another diffractometer. The latter two diffractometers shared one beam because only two horizontal beams were available in total for neutron scattering at that time. In Japan, they had to wait until 1964 to have the 10MW full power operation of JRR-2.

Among the many fields for which neutron scattering is utilized, magnetic problem is always one of the central issues because neutron scattering is almost the only source of the microscopic structural information from magnetic materials. At both JRR-2 and JRR-3 magnetic problems were taken up at first and since then many experiments have been done up to now. Another central issue is the structural research of the systems in which the heavy atoms and light ones coexist for which X-ray cannot give accurate information of the light atoms.

The experiments reported by JAERI at the International Symposium of Neutron Scattering held at Grenoble in 1963 typically reflected the above distinctive feature of the neutron scattering. Namely, magnetic structure of MnTe[4], Mn-Cr alloy, crystal structure of UF_4, frequency distribution in Ti-H[5] were reported there.

In those days, machines were not automatically operated as they are now done by computers. Scientists had to sit up at the machine side not to waste any neutrons while the reactor was running.

EARLY DAYS

The research activity was accelerated by the completion of JRR-3 in 1964. Both JRR-2 and JRR-3 had been used by the scientists of JAERI and universities. In 1965, JRR-3 became operational at full power. A TOF spectrometer of JAERI and a double-axis diffractometer shared by the Research Institute for Iron Steel and Other Metals of Tohoku University, Faculty of Science of Osaka University and JAERI were built there. Another diffractometer with polarized neutron capability was built there by ISSP.

When the starting period had passed and the initial constructions had settled down in JRR-2 it was well confirmed that the elastic scattering experiments can be performed steadily. Then there sprang up the next trial to challenge more difficult problems to measure inelastic scattering. As is well known, the cross section of the inelastic scattering caused by the excitations in solids like phonons or magnons is about three orders of magnitude smaller than that of elastic scattering but those information of lattice dynamics or spin dynamics are the really central problem of neutrons because X-ray cannot give rise to those information. All scientists were not necessarily confident that the flux at JRR-2 was high enough to measure inelastic scattering. Some people were feeling too pessimistic for the feasibility of phonon or magnon measurement at JRR-2. In 1965, however, the double-axis diffractometer of JAERI was changed into a triple-axis spectrometer of which type is now one of the most common neutron spectrometer to utilize the reactor neutrons. Then it was shown by the first successful measurement of inelastic scattering of the phonon dispersion relations of Cu-Ni alloy in Japan[6] that the flux at JRR-2 is high enough. Based on that experience phonons of CaF_2 was in estigated there by measuring not only the dispersion relation but also the intensity of the scattering and a new way of analysis of lattice dynamics was developed[7]. The first measurement of spin-waves in Japan was also performed with that spectrometer on MnTe[8] of which magnetic structure had been analyzed at JRR-2 long before that time[4]. This triple-axis spectrometer has been used as one of the main instruments of neutron scattering of JAERI group. Many experiments including phonon measurements of alloys related with martensitic transformations, measurements of magnons related with ferro-antiferromagnetic transitions, measurement of early stage of precipitation in alloys were performed with that instrument. Beam plug, monochromator shielding, operating system and many other part of this machine have been changed by now but it is still running at JRR-2.

At the university spectrometers in JRR-2 and JRR-3, studies of ferroelectricity, ionic crystals, magnetic problems of chromium, other metals and alloys were performed energetically.

In 1964 a time-of-flight spectrometer equipped with three-disc phased chopper was installed to HT-14 beam hole in JRR-2. This machine was used to measure paramagnetic scattering of MnO_2[9] and $KMnF_3$[10] and frequency distribution of metal hydrides. It is not easy, however, to maintain the high speed mechanical chopper in good condition to keep the phase precisely for a long operation time continuously. This machine was stopped years ago.

It was not until in 1971 that computers came to be used to operate the neutron spectrometers. The 8K word HITAC-10 computer was introduced by JAERI to operate both the triple-axis spectrometer and the chopper TOF machine with CAMAC simultaneously. Another HITAC-10 was introduced by Tohoku University to operate the triple-axis spectrometer. Computers were really expensive in those days.

THE PRIME OF JRR-2

As it turned out that JRR-2 is very powerful and the reactor operation became more stable, more and more experiments came to be performed there. It was unfortunate that the Electrical Communication Laboratory of Nippon Telegraph and Telephone Public Corporation stopped their neutron scattering activities early 60's though some structural studies of magnetic materials like MnAs etc. were performed in the very early days. This, however, gave the ISSP machine which used to share the beam hole more free utilization of the beam. The ISSP spectrometer was changed into a triple-axis spectrometer which has double monochromator for the first time in Japan. The spectrometer has been used by many university users for the studies of ferroelectricity, ionic crystals, superionic conductors, phase transition of low-dimensional magnetic materials, mixed magnetic materials with different anisotropy axis etc. The spectrometer is still now running at JRR-2.

In 1970, after ten years since the first spectrometer was installed in JRR-2 another conventional triple-axis spectrometer was installed in JRR-2 by the Faculty of Science of Tohoku University. Studies of magnetic excitations in metals and alloys in which magnetic electrons have strongly localized character to those with quite itinerant ones were performed intensively. In 1971, JAERI added a neutron topography machine in JRR-2. Studies of imperfections reflecting crystal growth, fundamental studies of neutron interferometry etc. were performed there. In 1975 JAERI added another triple-axis spectrometer with double monochromator in JRR-2. It was operated on the granite air-cushion floor for the first time in Japan. Studies of lattice dynamics of intercalated graphite, phase transition of alloys etc. were carried out with this machine. This machine is now moved to the end of T2 thermal guide of JRR-3M. In 1976, ISSP added a polarized neutron spectrometer. This machine was used not only for the standard measurement of magnetic studies but also as the prototype of the sophisticated Neutron Spectral Modulation(NSM) spectrometer proposed by Y.Ito. The last neutron scattering instrument installed to JRR-2 was the diffractometer with polarized neutrons which was installed by IMR of Tohoku University. Magnetic structure studies of uranium compounds, structure studies of metal hydrides were carried out with this diffractometer. It is now moved to T1 thermal guide in the guide hall of the JRR-3M reactor.

As described above, four triple-axis spectrometers, two polarized spectrometers, one TOF machine and one topography machine were once installed in JRR-2. Two spectrometers among them have been moved to the guide hall of JRR-3M. The rest six machines are still running at JRR-2.

JRR-3 IN THE PAST

In the early days of JRR-3, three spectrometers were installed in JRR-3 as described before. Later on a polarized neutron spectrometer was added by IMR of Tohoku University. This is the only machine which was once in old JRR-3 and again used in the JRR-3M. It is installed to the T1 thermal guide in the guide hall.

Since the flux of JRR-3 was not high enough to measure inelastic scattering, all of them have been used for elastic scattering. Studies of crystal structure of U_4O_9, magnetic structure of alpha-Mn, spin density in magnetic materials, measurement of magnetic form factor etc. were carried

out by making use of the old JRR-3 reactor. One of the unique experiments which were carried out there was an experiment to test feasibility of proton polarized filter as the neutron polarizer. To provide polarized neutrons, usually magnetic diffraction is used. In that case, however, the neutrons are monochromatized at the same time. Supermirror is another way to get polarized neutrons. It is very efficient for longer wavelength neutrons. On the other hand for rather short wavelength white beam as in the case of pulsed neutrons, the polarized proton filter is a good polarizer. The experiment was carried out by the collaboration of JAERI, IMR and High Energy Physics Institute(KEK). With the dynamic polarization of ethylene glycol, a satisfactory result that 80% polarization of neutrons was obtained[11].

PRESENT STATUS

The start of JRR-3M is a great epoch to the neutron scattering research in Japan. All the six horizontal beam holes in the reactor room are already being used. Four of them are used for the triple-axis spectrometers. Rest two are used for the high-resolution powder diffractometer and the neutron topography/optics machine. In the guide hall, six neutron scattering instruments have been installed up to now. They consists of two small-angle scattering machines, one triple-axis spectrometer, one neutron camera machine and two double-axis diffractometers.

Since the triple-axis spectrometers(TAS) have the most universal capability of thermal neutron scattering, it is quite natural that most of the beam holes with high intensity are occupied with them. Some of these triple-axis spectrometers are equipped with polarizer and polarization analyzer.

The high-resolution powder diffractometer(HRPD) equipped with sixty-four ^{3}He detectors in front of each of which are installed 6' fine collimators is the machine which is very powerful for the structural analysis especially for cases where the magnetic diffraction exists.

The neutron topography/optics machine is equipped with very delicate parts to control the goniometer angle with 0.01" increments. The whole apparatus is arranged on a vibration-free table enclosed in a double chamber which not only shields the background from the outside but also isolates the spectrometer from the reactor room environment to stabilize the temperature within less than 0.01 degree drift.

The two small-angle neutron scattering(SANS) machines in the guide hall are the first ones with large-scale two-dimensional position sensitive detectors installed in Japan. Among them, the one installed by JAERI at the end of 0.6nm guide is equipped with a 10m collimation guide and a 10m flight tube. The incoming neutron spectrum is chosen by the velocity selector made by KFKI Hungary which is installed between the end of the cold guide and the collimation guide. The $64x64cm^2$ ^{3}He position sensitive detector made by Riso Denmark can be moved inside the flight tube from 2 to 10m position from the sample. The flight tube can be turned on the air-cushion up to 15 degrees away from the direct beam axis. Another one built by ISSP at 0.4nm guide has 16m collimator and 16m fixed flight tube. It is equipped with Dornier velocity selector made in Germany and ORDELA two-dimensional detector made in USA.

The triple-axis spectrometer installed at the end of 0.2nm guide is the one moved from JRR-2 as described in the preceding section. Because it is installed at the end port it has great advantages that the scattering angle

can go up to very high, the monochromator can collect the beam from the total height of the guide and it can use rather short wavelength with low monochromator angle where no half-wavelength contaminatation comes. One of the two diffractometers installed by IMR at 0.2nm guide is also the one moved from JRR-2. This machine will be used mainly for single crystal samples. It will be added capabilities for triple-axis and four-circle functions in future. Another one is the diffractometer which used to be used at the old JRR-3 as the polarized neutron diffractometer. It will be used mainly for polycrystalline samples at the guide. It will be added the high-resolution powder diffractometer function in the future.

The neutron diffraction camera machine installed on the 0.2nm guide supplies monochromatic beam on the sample table on the air-cushion. This machine can be used not only as the camera but also for various purposes. Since the start of the JRR-3M in 1990, data are coming out of the machines already. The triple-axis spectrometers and the high-resolution powder diffractometer are running producing plenty of new results.

FUTURE

The neutron scattering instruments installed or to be installed at JRR-3M are summarized in Table 1 below. Twelve instruments have been installed up to now as described above. They are rather standard machines which are to be used on the well established way. Two double-axis diffractometers in the table will be modified in the future as described above.

Table 1 Neutron Scattering Instruments at JRR-3M

Instrument	Number		Location	Status	
Triple-axis spectrometers	8	4	Reactor room	4	running
		2	Thermal guide	1	running
		2	Cold guide		
High-resolution powder diffractometers	1	1	Reactor room	1	running
Double-axis diffractometers	2	2	Thermal guide	2	running
Small-angle scattering instruments	2	2	Cold guide	2	running
Topography, camera interferometry etc.	3	1	Reactor room	1	running
		2	Thermal guide	1	running
Special polarized neutron instruments(NSM,NSE)	2	2	Cold guide		
Other special neutron scattering instruments(TFS etc.)	2	1	Thermal guide		
		1	Cold guide		
Total	20			12	

In the coming few years, triple-axis spectrometers at thermal and cold guides, and other more challenging and sophisticated machines like neutron spectral modulation(NSM) machine, neutron spin-echo(NSE) machine, time focusing neutron spectrometer(NSF) and so on are planned in the guide hall.

In total, over twenty neutron scattering machines will be installed in JRR-3M[12], eight among which will be the triple-axis spectrometers and two are the small-angle scattering machines.

SUMMARY

Since the time when the first instrument was installed at JRR-2 the number of neutron scattering instruments increased gradually as shown in Fig.1 below. Most of them were triple-axis spectrometers in the past. It took nearly thirty years to have new JRR-3M in Japan. The circumstances of the neutron scattering research have been changed during these years. The rapid development of pulsed neutron sources affected the neutron scattering field greatly.

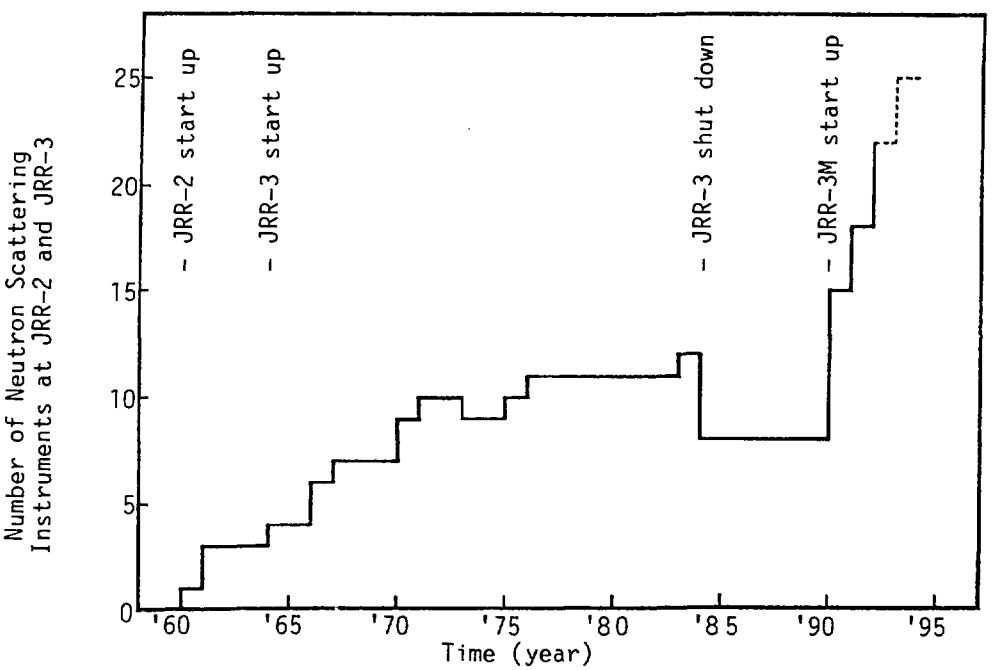

Fig.1 Time evolution of neutron scattering instruments at JRR-2 and JRR-3.

During these years, the field of the research to be studied with neutrons widened. In the early days, it was limited almost within magnetism, metallurgy and crystallography. Recently polymer became one of the most important fields of the neutron scattering research. Industrial application has just started.

The development is so quick recently. The pulsed neutrons are powerful for some problems. At the same time it is deeply recognized that the reactor neutrons are coming more and more indispensable.

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