19th meeting on Collaboration of Advanced Neutron Sources

March 8 – 12, 2010 Grindelwald, Switzerland

³He NEUTRON SPIN FILTER PROJECT IN JAPAN

Y. Sakaguchi, H. Kira, T. Oku, T. Shinohara, J. Suzuki, M. Nakamura, K. Suzuya, M. Arai, M. Takeda, S. Wakimoto, D. Yamazaki, S. Koizumi, Y. Endoh, K. Kakurai, *Japan Atomic Energy Agency, 2-4 Shirane, Shirakata, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan*

> Y. Arimoto, T. Ino, H. M. Shimizu, T. Kamiyama, High Energy Accelerator Research Organization, Oho 1-1, Tsukuba, Ibaraki 305-0801, Japan

K. Ohoyama, H. Hiraka, Institute for Materials Research, Tohoku University, Aoba-ku, Sendai 980-8577, Japan

K. Tsutsumi, Department of Physics, Tohoku University, Aoba-ku, Sendai 980-8578, Japan

K. Yamada

WPI Advanced Institute for Materials Research, Tohoku University, Aoba-ku, Sendai 980-8577, Japan

and

L-J. Chang

Nuclear Science and Technology Development Centre, National Tsing Hua University, Hsinchu, 30013, Taiwan

ABSTRACT

Polarized neutron scattering experiment is a powerful technique to separate magnetic scattering from nuclear scattering. However, it is a problem that scattered neutron intensity is weak in the experiment. The problem is expected to be solved by intensive pulsed neutron source of Japan Proton Accelerator Research Complex (J-PARC). To realize the experiment, it is necessary to introduce suitable neutron polarizers to J-PARC, in which pulsed neutrons in a wide energy range is used. ³He neutron spin filters are such devices. They polarize neutrons in a wide energy range and have large solid-angle. The devices can also be applied for a reactor-based facility in Japan, Japan Research Reactor No. 3 (JRR-3). In this paper, we report on our project of introducing ³He neutron spin filters into J-PARC and JRR-3. Our recent development on the glass cells for ³He spin filters is also reported.

1. Introduction

19th meeting on Collaboration of Advanced Neutron Sources

March 8 – 12, 2010 Grindelwald, Switzerland

Polarized neutron scattering experiment is a powerful technique to elucidate the nature of atomic magnetism, by separating magnetic scattering from nuclear scattering [1]. This technique can be applied for various research fields, such as magnetism, magnetic excitations, disordered magnetic systems, and surface/interface magnetism. Although it is a powerful technique, the problem is a weak scattered intensity. Since polarized neutrons are used, the intensity becomes half, at least, from incident unpolarized neutron beam. If we need to both polarize and analyze the neutron beam, the loss must be squared. The total scattered intensity will be much more reduced by a loss at neutron polarizers or related devices

In such a serious situation for performing polarized neutron scattering experiment, people hoped the appearance of intensive neutron source and the dream came true in Japan by a start of Japan Proton Accelerator-Research Complex (J-PARC). In several beam lines, it has been already demonstrated that the measurement time is shortened having better quality of spectra than previous measurements [2]. We can expect that the problem of weak scattered intensity in polarized neutron scattering experiment is solved by the intensive pulsed neutron source.

However, to realize the experiment, introduction of suitable neutron polarizers to J-PARC is necessary. In J-PARC, the available energy range of the white neutron source is wide. For instance, the available energy range in BL01 (4d-space Access Neutron Spectrometer: 4SEASONS) is between 5 and 500 meV (0.52 and 4.04 Å in wavelength) and that in BL14 (Cold-Neutron Disk-Chopper Spectrometer: AMATERAS) is between 1 and 80 meV (1.01 and 9.04 Å in wavelength). Therefore, it is desired to polarize all neutrons in the range. Among available neutron polarizers nowadays, magnetized Heusler crystals, Cu₂AlMn, are often used in diffractometers and triple-axis spectrometers. However, beam-mocochromatization is also made by this device and this is not suitable for the pulsed neutron source. Supermirrors are useful to cover wide energy range to polarize neutrons. However, the available energy range is limited in longer wavelength, say, longer than 2 Å. If the whole energy region including thermal neutrons like 1 Å is concerned, ³He spin filters, which are based on spin-dependent absorption, are useful. Although their operation is not simple compared to supermirrors and magnetized single crystals, the technique became mature and they are now operational at several institutions, such as Institute Laue-Langevin (ILL, France), National Institute of Standards and Technology (NIST, U. S. A.) and Forschungs-Neutronenquelle Heinz Maier-Leibnitz (FRM II, Germany). Marriage of the intensive pulsed neutron source and ³He neutron spin filter would yield us good future in polarized neutron scattering experiment. Once the device is introduced in J-PARC, the technique can also be utilized in a reactor-based facility in Japan, Japan Research Reactor No.3 (JRR-3).

In this paper, we report on our project of introducing ³He spin filters into J-PARC and JRR-3. To get higher ³He polarization and longer relaxation time, we investigate on optical system. This is presented in IP111. Our recent results of the ³He polarization test are presented in IP125. In addition, we report on the recent development of the glass cells in this paper.

2. Demand for Polarized ³He Neutron Spin Filters in Japan

2.1. Polarized ³He neutron spin filters in J-PARC

One of the great advantages of using pulsed neutrons is available high energy. For instance, there has been considerable attention on magnetic excitations in the field of high

19th meeting on Collaboration of Advanced Neutron Sources

March 8 – 12, 2010 Grindelwald, Switzerland

temperature superconductors and the presence of the magnetic excitations has been confirmed by the experiments using triple-axis spectrometers. As long as they use triple-axis spectrometers, the energy range was limited below 50 meV. However, using chopper spectrometer, MAPS in ISIS, the magnetic excitation has been measured by 200 meV and it was found that there is anomalous dispersion [3]. A complete separation of magnetic scattering from nuclear one would be required in such a field so that the origin of the scattering becomes clear. For this purpose, it is necessary to perform polarized neutron scattering experiment for high excitation energy and the use of ³He filters is the only solution to realize the experiment. The BL01 in J-PARC is the beam line for the inelastic neutron scattering to cover such high energy region (5 - 500 meV). Therefore, we are targeting this beam line to introduce ³He filters.

It is also an attractive point for ³He spin filters that they can have a large solid-angle. Indeed, we can cover wide angle range by aligning supermirrors as many as possible. In fact, there is such a beam line in ILL, which is D7. However, it would be more convenient and easier to install one ³He spin filter with a large solid-angle than aligning many supermirrors, although the operation of ³He spin filters is not so easy. Recently, ³He spin filters are often used in polarized neutron off-specular reflectivity. According to a recent report on a comparison between ³He spin filters and supermirrors, it was demonstrated that the data taken by ³He spin filter have better quality than supermirrors, without having artifacts and with low background [4]. In J-PARC, we are planning to construct a new reflectometer using polarized neutrons. In the reflectometer, we install two-dimensional detectors for off-specular neutron reflectivity measurement and grazing incidence small-angle neutron scattering measurement. To perform the polarized neutron reflectivity measurement, use of ³He spin filters will be required. Due to the same motivation, the introduction of ³He spin filters at smaller-angle neutron scattering beam line, which is under construction, is also planned.

One of the fascinating recent progresses in neutron imaging is neutron spin polarized imaging [5]. By polarizing and analyzing neutrons, magnetic fields can be visualized. To cover larger area for analyzing neutrons, the use of ³He spin filters is more convenient than supermirrors., Recently, we obtained neutron spin polarized imaging using supermirrors for analyzing neutrons on BL10 in J-PARC. It is worth trying to make neutron spin polarized imaging using ³He spin filters on the beam line.

To clarify a local magnetic order, it is necessary to make a complete separation of magnetic diffused scattering from nuclear one. Since the diffused scattering is weak, compared to sharp Bragg peaks, the experiment was not easy and the examples were few [6]. We expect that the combination of a powder diffractometer in J-PARC with ³He spin filters can solve this problem.

2.3. Polarized ³He neutron spin filters in JRR-3

Complementary use of a reactor to pulsed neutron source is necessary for our future development in neutron science. Once we could survey neutron scattering in a wide Q and ω range using pulsed neutrons, we can feed back the result into a reactor-base instrument, focusing on interesting range. In fact, ³He spin filters are often used in reactors, such as ILL, NIST and FRM-II. There are also demands to use ³He spin filters in JRR-3. A powder diffractometer, HERMES, can be utilized in various ways for polarized neutron scattering with ³He spin filters, such as magnetic form-factor studies, separation of magnetic and nuclear Bragg peaks, and determination of paramagnetic form factor. As well as BL01 in J-PARC, it is valuable to measure polarized inelastic scattering for high-energy excitations

19th meeting on Collaboration of Advanced Neutron Sources

March 8 – 12, 2010 Grindelwald, Switzerland

and we are interested in performing the experiments at a triple-axis spectrometer, TAS1, using ³He spin filters. Polarized neutron scattering technique can also be used for evaluating nuclear-spin incoherent scattering of hydrogen [1]. In some cases, there can be a large background of spin incoherent scattering of hydrogen in small-angle scattering region and there has been a strong demand to eliminate the background. We are planning to perform such experiment using ³He spin filters on SANS-J-II.

3. Research and Development of Glass Cells

There are several important factors to consider in the use of ³He neutron spin filters for polarized neutron scattering experiment. The cell production is one of the important factors. Firstly, it is desired to have large size of cells to cover incident or scattered neutrons as large as possible. Secondly, the quality of the window should be good enough to make optical pumping efficiently. Finally, the cell should keep ³He gas without making depolarization. The followings are our recent research and development on glass cells.

3.1. Production of large cells

In order to perform polarized neutron scattering experiment using ³He spin filters efficiently, it is desired to prepare large glass cells. To polarize ³He, we use spin-exchange optical pumping (SEOP). In this method, aluminosilicate glass named GE180 (G. E. Lighting Component Sales) is supposed to be the best glass for ³He spin filters. But the fabrication of the glasses is not so easy; narrow operation temperature range, viscous, thermal strain is easy to be made. In addition, only one size of the tube, 15mm in diameter, is available on the market. This makes more difficult to produce large cells with GE180. At some most experienced area, like a group in NIST, they have already prepared large cells with 100 mm-windows in diameter [7]. We have produced the glass cells with 40 mm in diameter. But we are trying to produce larger cells.

We prepared glass cells with 50 mm in diameter, and 70 mm in length, as shown in Fig.1(a). When we produce the cells, we made careful annealing at 800 °C. Thermal strain can be observed visually, by using an optical strain gauge, in which two linear polarizers are set. No colour change means no strain on the glass, while colour change means a presence of a strain on the glass. Fig 1(b) shows that the thermal strain is almost eliminated, due to the effect of annealing. We also performed pressure test. The three cells endured pressure of 6 kg/cm². For one of the cells, we applied more pressures and it was broken at 8 kg/cm².

To pursue the possibilities of producing much larger GE180 glass cell, we prepared GE180 glass plates. Construction of large rectangular cells, or cylindrical cells with large window might be possible if the components of GE180 glass plates could be prepared. As seen in Fig.2(a), GE180 glass plates with 50 mm in diameter and 3 mm in thickness were produced. But there are bubbles in the plates. Also, a strain remained (Fig.2(b)). We are still under study.

ICANS XIX, 19th meeting on Collaboration of Advanced Neutron Sources

March 8 – 12, 2010 Grindelwald, Switzerland

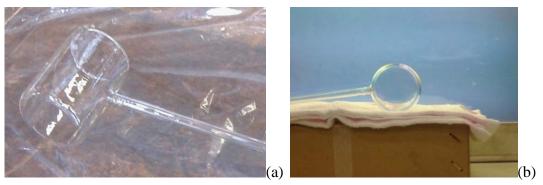


Fig.1 (a) GE180 glass cell with 50 mm in diameter and 70 mm in length. (b) Image of the cell observed by an optical strain gauge.

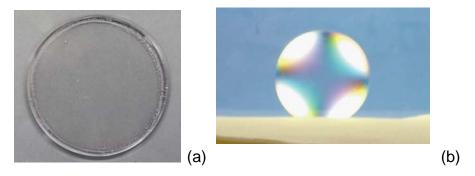


Fig.2(a) GE180 glass plate with 50 mm in diameter and 3 mm in thickness, (b) Image of the GE180 glass plate observed by an optical strain gauge.

3.2. Glass window

The quality of the window is also important, because circular polarized laser beam passes through the glass window before it illuminates rubidium gas. The loss of the intensity and the loss of the circular polarization result in less efficiency in the optical pumping. We measured circular polarization of the transmitted laser beam for a window as shown in Fig. 3(a). At the centre, it is almost unity, while it is about 0.8 near the edge region. To use the laser intensity to the maximum, this reduction is desired to be restored. Since the reduction is supposed to be caused by structural distortion, it would be improved by thermal annealing. Therefore, we annealed the cell, heating it by 800 °C. After that, the circular polarization restored by almost unity and the strain was not detected in the image of the optical strain gauge (Fig.4). This would give an improvement in the efficiency of the optical pumping.

ICANS XIX, 19th meeting on Collaboration of Advanced Neutron Sources

March 8 – 12, 2010 Grindelwald, Switzerland

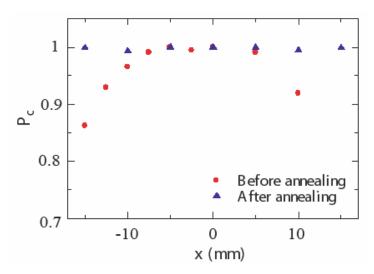


Fig. 3(a) Circular polarization, P_c , of the laser beam which passed through a cell window, as function of the position from the centre on the window.

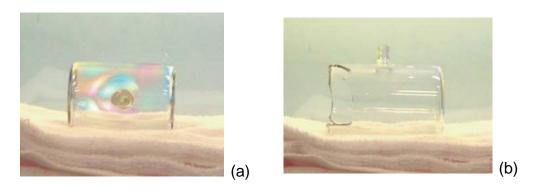


Fig.4 Image of GE180 cell observed by an optical strain gauge (a) the cell without annealing (b) the cell after being annealed at $800\,^{\circ}$ C.

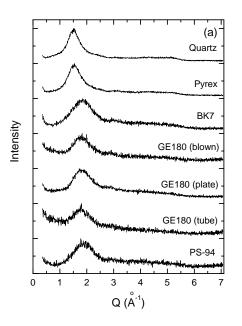
3.3. Structural Characterization

Since helium atoms are smaller than other inert gas such as argon, the permeation can be a problem in a certain type of glasses [8]. For instance, quartz is made of silicon and oxygen atoms and they are strongly bonded each other with covalent bonds. The Si(O_{1/2})₄ tetrahedral units are supposed to be completed, forming network. However, they are loosely packed when glasses are formed, and there are microscopic "holes" in the glasses. From this reason, a leakage of gasses can occur in a quartz container. The leakage can be a problem to maintain helium gas in the glasse cell for ³He spin filter. Furthermore, the permeation of helium gas in the glasses causes more collision and hence, more chance for depolarization. In the production of vacuum containers, to avoid such leakage, addition of glass modifier, such as alkali atoms and boron atoms, is often made. The modifier breaks the network, and at the same time, it fills the holes. As long as we use the glasses for neutron scattering, boron is not a suitable element as additive, because it has a large absorption cross section. Also, we need to consider ease of glass fabrication. GE180 is the

19th meeting on Collaboration of Advanced Neutron Sources

March 8 - 12, 2010 Grindelwald, Switzerland

easiest glass for fabrication among aluminosilicate glasses. But it is not easy yet to produce large size of the cells as we described above. Exploration of substitutional glasses might be needed. Moreover, it has been pointed out that the glasses should completely be blown in the process of the cell production to get a long relaxation time [7]. This means that the difference of the structure, which is caused by a thermal treatment, can affect the quality of spin filters. From such backgrounds, we had an interest in the microscopic structural characterization, which can be a good measure of the quality of the cells.



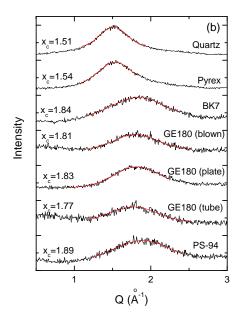


Fig.5(a) X-ray diffraction curves for several glasses. For GE180 glasses, three types of samples were measured; completely blown glasses (blown), the plate, shown in Fig.2 (plate), and original glass tube (tube). (b) enlarged diffraction pattern in the Q-range between 0.5 and 3.0 A-1. The x_c indicates the centre of the Gaussian fit for the first peak.

Fig.5 shows the result of X-ray diffraction (XRD) measurements for several glasses. In the diffraction curve of quartz, there is a clear broad peak around 1.5 Å ⁻¹. This is called First Sharp Diffraction Peak, and indicates medium-range order (MRO). The peak is interpreted as an indicative peak of either the cages formed by the topological connection of the tetrahedral units [9] or void [10]. Therefore, the peak might be a measure of the size of the microscopic holes. The peak for GE180 is located at higher-Q than that for quartz. This indicates that the size of the hole in GE180 is smaller than that in quartz. In fact, GE180 is less permeable to helium than quartz. There is a small difference in the peak position among three types of GE180 glasses. In addition, there is a small peak at 3 Å ⁻¹ in the diffraction curve for the blown GE180 glasses, while it seems that there is no such peak in the curve for original glass tube. These differences might indicate the difference in the structure depending on a thermal treatment. We also measured XRD for PS-94 (Nippon Electric Glass Co., Ltd.), which is lead-free glass. This does not contain boron and therefore, we thought that this can be a candidate of substitutional glasses for GE180. As far as we see the peak position, this seems to work as ³He spin filters. However, after

19th meeting on Collaboration of Advanced Neutron Sources

March 8 – 12, 2010 Grindelwald, Switzerland

some trials of glass working, it turned out that it has high thermal expansion coefficient and its fabrication can be more difficult.

4. Summary

We are preparing ³He neutron spin filters to install them on the beam lines in both J-PARC and JRR-3. To get higher ³He polarization and longer relaxation time, we investigate on the optical system (IP111) and glass cells (this paper) and perform ³He polarization test (IP125). We promote this project to open up a new stage in polarized neutron scattering studies. We believe that its realization will yield a new insight in diversified research fields such as magnetism, strongly correlated electron systems, high temperature superconductivity, spintronics, polymer science, and life science.

5. Acknowledgements

This research project has been supported by Quantum Beam Technology Project, JST, MEXT. We thank Dr. Soyama and Dr. Maruyama for assisting in the X-ray diffraction measurements.

6. References

- 1. G. W. Williams, *Polarized Neutrons* (Clarendon Press, Oxford, 1988).
- 2. M. Nakamura, R. Kajimoto, Y. Inamura, F. Mizuno, M. Fujita, T. Yokoo and M. Arai, *J. Phys. Soc. Jpn.* **78** (2009) 093002.
- 3. J. M. Tranquada, H. Woo, T. G. Perring, H. Goka, G. D. Gu, G. Xu, M. Fujia and K. Yamada, *Nature*. **429** (2004) 534.
- 4. A. K. Petoukhov, K. H. Andersen, D. Jullien, E. Babcock, J. Chastagnier, R. Chung, H. Humblot, E. Lelièvre-Berna, F. Tasset, F. Radu, M. Wolff and H. Zabel, *Physica*. **B 385-386** (2006) 1146.
- 5. Y. Wu, W. Dmowski, T. Egami and M. E. Chen, J. Appl. Phys. **61** (1987) 3219.
- 6. N. Kardjilov, I. Manke, M. Strobl, A. Hilger, W. Treimer, M. Meissner, T. Krist and J. Banhart, *Nature Physcs.* **4** (2008) 399.
- 7. D. R. Rich, T. R. Gentile, T. B. Smith, A. K. Thompson and G. L. Jones, *Appl. Phys. Lett.* **80** (2002) 2210.
- 8. F. J. Norton, J. Am. Ceram. Soc. 36 (1953) 90.
- 9. A. C. Wright, J. Non-Cryst. Solids 179 (1994) 84.
- 10. S. R. Elliott, Phys. Rev. Lett. 67 (1991) 711.