ELSEVIER
CrossMark
Available online at www.sciencedirect.com
ScienceDirect
Physics Procedia 60 (2014) 167 – 174
Physics Procedia

Union of Compact Accelerator-Driven Neutron Sources (UCANS) III & IV

Activity of Hokkaido University Neutron Source, HUNS

Michihiro Furusaka[a,*], Hirotaka Sato[a], Takashi Kamiyama[a], Masato Ohnuma[a] and Yoshiaki Kiyanagi[a,b]

[a]*Faculty of Engineering, Hokkaido University,Kita 13, Nishi 8, Kita-ku, Sapporo, Hokkaido 060-8628, Japan*
[b]*Present address: Graduate School of Engineering, Nagoya University, Furo cho, Chikusa-ku, Nagoya, Aichi 464-8603, Japan*

Abstract

Hokkaido University neutron source, HUNS was completed in 1973, and has been used actively for developments of moderators, neutron instruments, neutron devices and new methods for 40 years although its power is not so high. Recently, a pulsed neutron imaging method has been developed and a new type of small angle neutron scattering method has been also developed. The pulsed neutron imaging is a unique method that can give the physical quantities such as crystallographic quantities of materials over wide area of the real space. So far, the small angle neutron scattering (SANS) is considered to be impossible at a neutron source with a power of HUNS. However, mini focusing SANS (mfSANS) was developed and proved to be useful. Here, we present the present activities on the pulsed neutron imaging and mfSANS at HUNS.

Peer-review under responsibility of the Organizing Committee of UCANS III and UCANS IV

Keywords: HUNS; Pulsed neutron imaging; mfSANS.

1. Introduction

The construction of 45 MeV Hokkaido University electron linac (HU Linac) was started in 1971, and reached routine operational mode in full energy from the 1973-1974 period. The maximum electron energy is 45 MeV and usual operation energy is 35 MeV. The electron burst width can be changed from 10 ns ~ 3 μs, and the repetition rate from single to 200 pps. Now the highest repetition rate is 50 pps to elongate the life of an electron gun. HU Linac has been used for many purposes, like neutron science, pulse radiolysis for chemical applications, electron beam monitoring, parametric X-ray study and in the recent years it was proved that soft error test experiments could be carried out at this facility. As a facility that is owned and run by the faculty of engineering, so many students have been educated in this environment and there have been very active collaborations with many other institutions and companies, domestic as well as international ones.

* Corresponding author. Tel.: +81-11-706-6677; fax: +81-11-706-6677.
E-mail address: furusaka@eng.hokudai.ac.jp

Peer-review under responsibility of the Organizing Committee of UCANS III and UCANS IV
doi:10.1016/j.phpro.2014.11.024

Hokkaido University neutron source (HUNS) was completed as a compact accelerator driven neutron source (CANS) till 1973. The electron linac was the neutron generator and a cold neutron source was equipped firstly in the world at the accelerator driven neutron sources. The neutron yield is about 10^{12} n/sec at 1 kW operation. The neutron source was used actively for 40 years for various neutron fields. First, it was used to study cold neutron moderators, and a 20 K solid methane cold moderator was developed [1], which has the best neutronic performance even now at CANS. Furthermore, with use of HUNS a quasi-elastic neutron spectrometer was developed [2]. These techniques were transferred to KENS (High Energy Accelerator Research Organization Neutron Source), and contributed to high performance neutron scattering experiments at KENS. After then new moderator development was accomplished for the J-PARC neutron source. The developed cold neutron source, the coupled hydrogen moderator [3], was used not only at J-PARC but also at SNS in the USA. As a neutron device a neutron magnetic lens was examined firstly in the world and neutron focusing effect was clearly demonstrated [4]. This result led to the intensive study of the neutron devices in Japan.

Recently, pulsed neutron imaging is being studied. Neutron imaging is one of the useful radiation transmission imaging, methods and usually uses neutrons with a wide energy range. However, by using the pulsed neutron source we can obtain transmission spectra at each pixel of a two-dimensional position sensitive detector coupled with the time-of-flight method. The spectra reflect structure of the neutron total cross section, in which there exists crystallographic information, elemental information, magnetic field information and so on [5]. We use Bragg edge transmission, polarized neutron for magnetic field imaging, and resonance transmission for elemental analysis and temperature measurement. The method is considered to be very useful for material evaluation and other nondestructive inspection.

Neutron scattering experiments such as diffraction, quasi-elastic neutron scattering were performed at HUNS, but they were quitted after KENS operation. The recent development of the neutron devices has opened new field in the instruments. The small angle neutron scattering using a focusing device is one of the prominent examples of the new instruments suited for the CANS. This idea makes the SANS instrument very compact (min focusing SANS: mfSANS) and it has been proved to give useful data even at a CANS like HUNS.

These new developments are based on HUNS and have given impact on the CANS applications. Therefore, here we introduce the recent activities on SANS and pulsed neutron imaging.

2. Small/medium-angle neuron scattering

The ratio of neutron flux at large facilities to the one at CANS is quite large, of the order of 10^3-10^5. Despite of the fact, we believe that CANS is still competitive in many important areas. The key to the competitiveness is “taylor-made”, like tailor-made accelerator, moderator, instrument and detector. Let us start with one example, small/medium-angle neutron scattering.

In case of metal material development, characterization method is quite important. What you can develop is depending on the characterization method. Down to about a few nm in size, the most important characterization method is transmission electron microscope, TEM. Nowadays, material developers are more and more interested in very fine structures, of the order of nm in matters. In such length scale, TEM cannot see such structures and even if they were detected it was not so straight forward to obtain overall, statistically correctly averaged information over all the sample volume. Small-angle X-ray and neutron scattering methods are inherently strong in this area and can fill the area that TEM is not appropriate.

The *Q*-range necessary for this kind of measurement is centered around 1 nm^{-1} region, therefore we have chosen the range of 0.2 to 5 nm^{-1}. If we can concentrate in measuring this region, intensity gain compared with measuring conventional SANS region is around 10^2-10^3. Of course we cannot conduct same experiment at CANS like at large facilities, but when we focus on very necessary information for specific characterization, we can design an instrument appropriate for the specific length scale. The Intermediate-Angle Neutron Scattering instrument, iANS, is one of such instruments. It focuses to the nanoscopic structure studies in metal materials.

In Fig. 1, SANS in steel samples with and without nanoscopic precipitates are shown. Small/medium-angle neutron scattering was measured with good enough statistics in the Q-range of 0.2 to 5 nm^{-1} with the measuring time of about 6 hours. Of course Q below 0.2 nm^{-1} cannot be measured by this instrument as expected.

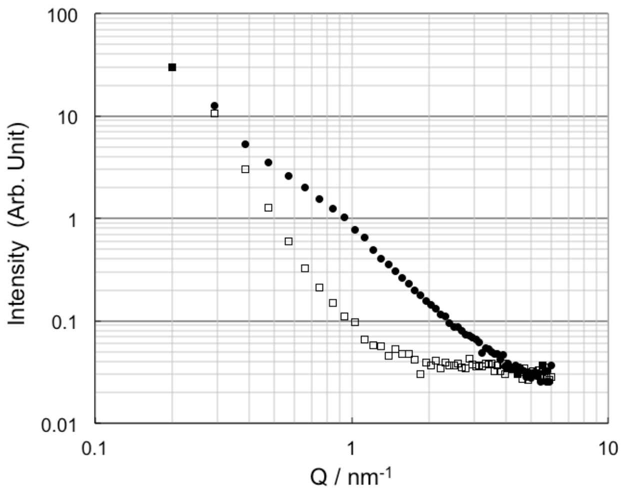

Fig. 1. SANS in steel samples with (filled markers) and without (open markers) nanoscopic precipitates.

The design of iANS is as the followings: by using a natural collimation defined by the moderator (120 mm) and sample size (14 mm), the incident collimation is about 10 mrad using about 5 m of incident flight path. The scattered path length is also quite short, about 460 mm, far less than those of conventional small-angle neutron scattering instruments. Because this instrument uses time-of-flight (TOF) method, it can utilize a good wide wavelength band, from about 0.1 nm and up to about 1.5 nm. By utilizing such wide lambda-range, it can measure minimum *Q* of around 0.2-0.3 nm^{-1}, and a Bragg-diffraction region simultaneously.

The half-inch diameter linear position sensitive detector tubes (LPSD) made by General Electric (GE) are used. As a beam stop, a piece of cadmium plate is put at the center of one of the LPSD tubes. The beam stop has small pinholes such that very small portion of direct beam can pass through it and transmission can be measured simultaneously with small/wide-angle scattering.

Instead of using a vacuum chamber for the scattered path, a helium gas chamber is put in between the sample and the detector. A proper vacuum chamber is also under development.

3. New developments for pulsed neutron imaging

3.1. Crystallographic structure analysis

The pulsed neutron Bragg-edge transmission imaging is expected to be a new crystallographic/metallographic analysis tool that can obtain different information from SEM-EBSD, X-ray/neutron diffraction and synchrotron radiation microtomography. This is because this method can non-destructively visualize bulk material information over large area with reasonable position resolution. The key technologies, 2D position-sensitive neutron detectors and data analysis software, have been developed at HUNS, and they were exported to J-PARC [5-9]. As new 2D detectors, a wavelength-shifting fiber detector [10], direct-readout pixel-type detectors [11, 12], a boron-type GEM detector [13] and neutron image intensifier [14] were developed/tested at HUNS. Now, in this section, we present recent progress on a data analysis software and application activities of the Bragg-edge transmission imaging at HUNS.

HUNS is a compact accelerator-based short-pulsed cold neutron source. A coupled moderator is usually used to increase neutron flux at sample/detector position. For this reason, the wavelength resolution is not so good, about less than 3% for 0.4 nm wavelength neutrons. Thus, it is not so easy to observe shift of a Bragg "edge" caused by strain. On the other hand, it is relatively easy to observe shape change of a Bragg-edge transmission spectrum caused by change of crystal orientation distribution (texture), and also increase of transmission intensities due to the primary extinction effect (multiple diffraction inside a crystallite) caused by coarse crystallites. Therefore, through pulsed neutron transmission experiments at HUNS, we developed a data analysis software, RITS (Rietveld Imaging of Transmission Spectra), for both investigation of reasons of the shape/intensity change and quantitative estimation of texture/crystallite size [15]. Fig. 2 shows Bragg-edge transmission spectra of various α-irons measured at HUNS, and the profile fitting curves obtained by the RITS code. The changes of shape/intensity due to texture/crystallite size are experimentally observed. Furthermore, transmission spectra calculated by the RITS code follow experimental data with good agreement. Through such profile fitting analyses with the RITS code, we successfully evaluated material parameters on texture (preferred orientation and degree of crystallographic anisotropy) and microstructure (crystallite size) at HUNS. Then, the RITS code was continuously improved, and was also adapted to strain analysis [5, 8] and crystalline phase analysis [16], experimentally performed at J-PARC MLF.

So far, various applications of the Bragg-edge transmission imaging are also in progress at HUNS; for example, qualitative observation/imaging of phase transformation of Pb [17] and LBE (Pb-Bi eutectic) [18]. As examples using the RITS code, quantitative texture/microstructure imaging of welded α-iron plates [15], cultural heritage such as a Japanese sword, a Mg alloy and a quenched steel rod etc. have been already performed. It is expected that such application activities at HUNS/CANS become more active in near future owing to new development of neutron imaging detectors and data analysis codes.

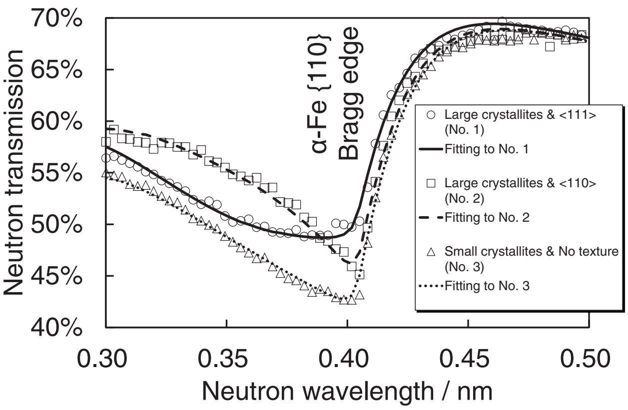

Fig. 2. Bragg-edge transmission spectra measured at HUNS, and the profile fitting curves obtained by RITS.

3.2. Neutron resonance absorption spectroscopy

Neutron resonance absorption spectroscopy (N-RAS) using the neutron time-of-flight (TOF) technique is a technique to distinguish the dynamics of individual nuclides with resonance peaks on epithermal neutron region. The technique makes it possible to observe a motion of a particular nuclide with measuring the neutron resonance absorption spectrum. In the measured spectrum, the line width of the peak is affected by the thermal motion of the target nuclide. This ability makes N-RAS as a unique material research technique of an individual observation of plural nuclide [19, 20].

One of the advantages of N-RAS is higher sensitivity than the normal neutron scattering spectroscopy since resonance absorption cross sections take sometimes large values. Then it was considered as suitable for a compact neutron source such as at Hokkaido University 45 MeV electron linac facility (HUNS). The new N-RAS spectrometer at HUNS has the arrangements of counting prompt gamma-rays emitted from nuclei absorbing neutrons or recording neutron absorption directly as a function of neutron flight time. The N-RAS spectrometer is set up at a light water moderator [21, 22]. The flight path length from the moderator to a sample is variable, but normally around 13.3 m. A vacuumed neutron beam flight path has two B_4C collimators, which can be used to control the size and the position of the neutron beam spot on the sample.

The sample absorbs neutrons with a resonance energy E_R and then emit the prompt gamma-ray by (n, γ) reaction. For neutron absorption spectrum recording, Li-glass scintillator pixel-type detectors are set up at behind of the sample. For the prompt gamma-ray recording, eighteen BaF_2 scintillation detectors are installed both sides of the sample and the events are recorded by time analyzers. The counter banks are shielded for neutrons and gamma-rays by Fe, B and Pb. Whole spectrometer is equipped on a large turntable for the adjustment of an alignment of the computer tomography (CT) measurement. The CT stage has two motions, one is sample rotation and the other is neutron slit movement. The neutron slit, which is made by B_4C, can select the appropriate width form 0 to 3 mm. All stage motion can control remotely.

Fig. 3 shows experimental number density (N_d) distribution results with N-RAS with CT for a cylindrical cell. The qualitative N_d value was taken by fitting of the resonance absorption equation with the neutron pulse shape effect for 4.28 eV for $_{73}$Ta and 1.45 eV for $_{49}$In, respectively. The analyzed N_d spatial distributions of Ta and In are shown as the tomograms in Fig. 3 upper. The $_{73}$Ta and $_{49}$In regions in the cross section on the sample cell are all bright in the figure; the result agrees with the actual object structure. The lower figures are density distributions along the center cross sections of the upper figures. Each distribution shows the actual density distributions (green shaded areas) within the 10% difference. These reconstructed results can be said that N-RAS/CT is possible to make the quantitative CT measurement even in the compact neutron facility.

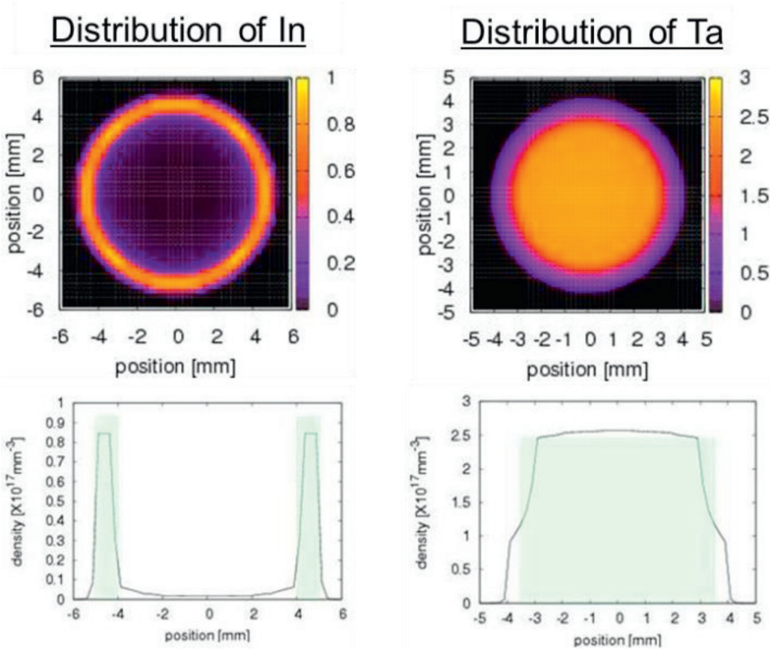

Fig. 3. The results of the CT reconstruction for Ta (left) and In (right) distributions inside the sample cell. In the upper tomogram the bright area corresponds to the Ta or In existence; Lower corresponds to their density distributions at center cross.

3.3 Fundamental experiments for magnetic field imaging

By using polarized neutron we can obtain the spatial distribution of magnetic field [23,24]. The magnetic field is evaluated by measuring the rotation of a neutron spin as the change of the neutron polarization after passing the magnetic field. With use of a 2-dimensional position sensitive detector coupled with the time-of-flight method we can map the spatial dependent magnetic field. The neutron spin rotation angle φ due to the Larmor precession in a magnetic field B is expressed by following equation,

$$\varphi = \frac{\gamma}{v}\int_{path} B ds \qquad (1)$$

where γ is the gyromagnetic ratio of the neutron and v the neutron velocity. Therefore, a path integrated B value is measured from the information of φ. One of unique characteristics of the pulsed neutron method is that one can evaluate more easily than the steady sources the absolute value of the magnetic field. So as to check the feasibility of the quantitative evaluation we have performed a simple coil measurement. The experimental setup is shown in Fig. 4.

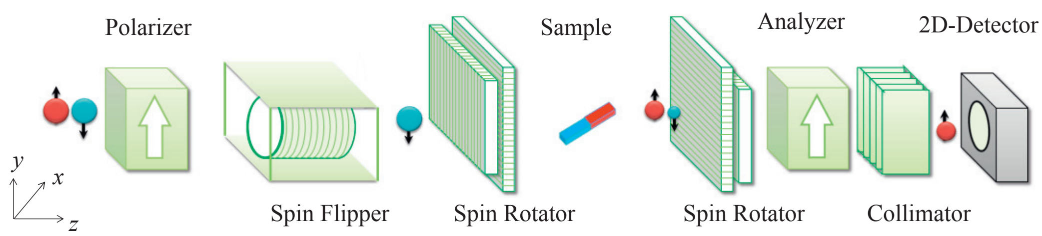

Fig. 4. Experimental setup of magnetic field measurement.

Polarizer and analyzer are stacked bent supermirrors. The detector is 5inch-RPMT with ZnS(Ag)/^{6}LiF and its pixel size is 1 mm. The flight path length is 7.78 m, and the beam size is 10 mm (W) and 20 mm (H). Measuring time was 30 min per measurement. The magnetic field was measured by a Hall probe, and the value was 3.56 mT at a current of 1.5 A. Fig. 5 shows the polarization as a function of neutron wavelength at three different orientations. The polarization was expressed by following equation. Here, λ is the neutron wavelength, and n_i is direction cosine of the magnetic field, and i corresponds to x, y and z.

$$P_i / P_0 = 1 - (1 - n_i^2)(1 - \cos\varphi) \qquad (2)$$

$$\varphi = \omega_P \lambda \qquad (3) \quad (\omega_P : Polarization\ frequency)$$

The period of each direction is almost the same as shown. The magnetic field evaluated by polarizer neutron transmission was 3.636 ± 0.032 mT. The value is within 95% of the Hall probe value.

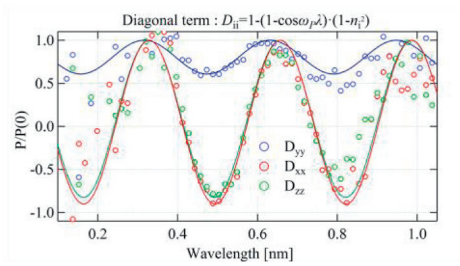

Fig. 5. Polarization of the three directions.

4. Concluding remarks

CANS's have been around for long time, since 1960's. The use of such CANS's have been mainly on R&D's of neutron sources, instruments, devices and not much about real scientific researches or industrial applications. We showed in these few years that even with a CANS like HUNS, small/medium-angle neutron scattering studies can be carried out and it even has advantages compared with larger facilities; with an in-house neutron source, timely measurements can be done, which is essential for characterization study of new material development; there is no limitation in trying new concept or new materials. Of course, once crudely information is obtained by a CANS, larger facilities would be much better to explore more in details.

Taylor made source and instrument is a key to such future CANS instruments. We are just at the beginning of the development of taylor made CANS's and instruments that are competitive with larger-facility instruments. There are many possibilities of developing accelerators tailor-made to specific applications, such as the energy, pulse width, pulse repetition rate, beam emittance. One example would be a coded pulse structure accelerator or sinusoidally intensity modulated pulse sequence accelerator. Neutron source should also be tailored. We could not employ a reentrant-hole type moderator on larger facilities because such a moderator should be shared with several instruments, but at a CANS, there would be a possibility of using such a moderator for single purpose instrument.

The Bragg-edge imaging method was developed at HUNS and a new instrument is under construction for the J-PARC. However, if we could tailor-made the accelerator, neutron source and instrument, CANS would be still competitive. For example short-pulsed beam from an accelerator delivered to very short pulse decoupled moderator coupled with a high-Q_c guide tube would be a great combination for a Bragg-edge imaging instrument. With the short pulse, we are able to increase pulse repetition rate without sacrificing neutron wavelength band-width. Also, with a tightly coupled guide tube, we can increase beam divergence and large gain in intensity without sacrificing Bragg-edge profile resolution. It should be noted that the maximum counting rate of a detector system for such measurement is often the limiting factor of the maximum statistics.

We are just at the beginning of a new era opened by unseen new CANS's. We have not exploited new low-emittance neutron source such as the reentrant-hall moderator coupled with new accelerator technologies. We have not yet asked to accelerator developers that what kind of accelerator would be best for a CANS for a specific application. We will need almost same numbers of CANS systems as the numbers of applications in future.

Acknowledgements

The authors thank Dr. Shinohara at J-PARC Center for the collaboration in the magnetic field measurement. This work partially include the results by a Grant-in-Aid for Scientific Research (S) from Japan Society for the Promotion of Science (No. 23226018), and also the result of research of analysis technique for a MOX fuel pellet by neutron resonance absorption entrusted to Hokkaido University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

References

[1] K. Inoue, Y. Kiyanagi and H. Iwasa, Nucl. Instrum. Methods **192** (1982) 129.
[2] K. Inoue, Y. Kiyanagi, H. Iwasa and Y. Sakamoto, Nucl. Instrum. Methods **178** (1980) 459.
[3] Y. Kiyanagi, N. Watanabe and H. Iwasa, Nucl. Instrum. Methods A **312** (1992) 561.
[4] H. M. Shimizu, H. Kato, T. Oku, Y. Suda, Y. Ogawa, H. Iwasa, T. Kamiyama, Y. Kiyanagi and T. Wakabayashi, Physica B **241-243** (1998) 172.
[5] Y. Kiyanagi, H. Sato, T. Kamiyama and T. Shinohara, J. Phys. Conf. Ser. **340** (2012) 012010.
[6] H. Sato, O. Takada, K. Iwase, T. Kamiyama and Y. Kiyanagi, J. Phys. Conf. Ser. **251** (2010) 012070.
[7] K. Iwase, H. Sato, K. Mori, T. Kamiyama, T. Ishigaki and Y. Kiyanagi, Metall. Mater. Trans. A **42** (2011) 2296.
[8] K. Iwase, H. Sato, S. Harjo, T. Kamiyama, T. Ito, S. Takata, K. Aizawa and Y. Kiyanagi, J. Appl. Crystallogr. **45** (2012) 113.
[9] Y. Kiyanagi, T. Shinohara, T. Kai, T. Kamiyama, H. Sato, K. Kino, K. Aizawa, M. Arai, M. Harada, K. Sakai, K. Oikawa, M. Ooi, F. Maekawa, H. Iikura, T. Sakai, M. Matsubayashi, M. Segawa and M. Kureta, Phys. Proc. **43** (2013) 92.
[10] Y. Kiyanagi, N. Sakamoto, H. Iwasa, T. Kamiyama, F. Hiraga, S. Sato, H. Sagehashi, T. Ino, M. Furusaka, J. Suzuki, A. Gorin, I. Manuilov, A. Ryazantsev, K. Kuroda, K. Sakai, F. Tokanai, H. Miyasaka, T. Adachi, T. Oku, K. Ikeda, S. Suzuki, K. Morimoto and H. M. Shimizu, IEEE Trans. Nucl. Sci. **52** (2005) 371.
[11] Y. Kiyanagi, K. Mizukami, T. Kamiyama, F. Hiraga and H. Iwasa, Nucl. Instrum. Methods A **542** (2005) 316.
[12] H. Sato, O. Takada, S. Satoh, T. Kamiyama and Y. Kiyanagi, Nucl. Instrum. Methods A **623** (2010) 597.
[13] S. Uno, T. Uchida, M. Sekimoto, T. Murakami, K. Miyama, M. Shoji, E. Nakano, T. Koike K. Morita, H. Sato, T. Kamiyama and Y. Kiyanagi, Phys. Proc. **37** (2012) 600.
[14] T. Kamiyama and Y. Kiyanagi, Phys. Proc. **26** (2012) 231.
[15] H. Sato, T. Kamiyama and Y. Kiyanagi, Mater. Trans. **52** (2011) 1294.
[16] H. Sato, T. Shinohara, R. Kiyanagi, K. Aizawa, M. Ooi, M. Harada, K. Oikawa, F. Maekawa, K. Iwase, T. Kamiyama and Y. Kiyanagi, Phys. Proc. **43** (2013) 186.
[17] Y. Kiyanagi, T. Kamiyama, T. Nagata, F. Hiraga and S. Suzuki, Physica B **385-386** (2006) 930.
[18] O. Takada, T. Kamiyama and Y. Kiyanagi, J. Nucl. Mater. **398** (2010) 129.
[19] K. Kaneko, T. Kamiyama, Y. Kiyanagi, T. Sakuma and S. Ikeda, J. Phys. Chem. Solids **60** (1999) 1499.
[20] K. Tokuda, T. Kamiyama, Y. Kiyanagi, R. Moreh and S. Ikeda, Jpn. J. Appl. Phys. **40** (2001) 1.
[21] T. Kamiyama, N. Miyamoto, S. Tomioka and T. Kozaki, Nucl. Instrum. Methods A **605** (2009) 91.
[22] T. Kamiyama, N. Miyamoto, S. Tomioka, T. Kozaki, 2009 IEEE Nucl. Sci. Symp. Conf. Rec., 10.1109/NSSMIC.2009.5402369 (2009) 1266.
[23] N. Kardjilov, I. Manke, M. Strobl, A. Hilger, W. Treimer, M. Meissner, T. Krist, J. Banhart, Nat. Phys. **4** (2008) 399.
[24] T. Shinohara, K.Sakai, M.Ohi, T.Kai, M.Harada , K.Oikawa , F.Maekawa, J.Suzuki, T.Oku, S. Takata, K.Aizawa, M.Arai, Y.Kiyanagi, Nucl. Instrum. Methods A **651** (2011) 121.