

| Title | Energy resolution of pulsed neutron beam provided by the ANNRI beamline at the J-PARC/MLF | |
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| Author(s) | Kino, K.; Furusaka, M.; Hiraga, F.; Kamiyama, T.; Kiyanagi, Y.; Furutaka, K.; Goko, S.; Hara, K. Y.; Harada, H.; Harada, M.; Hirose, K.; Kai, T.; Kimura, A.; Kin, T.; Kitatani, F.; Koizumi, M.; Maekawa, F.; Meigo, S.; Nakamura, S.; Ooi, M.; Ohta, M.; Oshima, M.; Toh, Y.; Igashira, M.; Katabuchi, T.; Mizumoto, M.; Hori, J. | |
| Citation | Nuclear instruments & methods in physics research section a-accelerators spectrometers detectors and associated equipment, 736, 66-74 https://doi.org/10.1016/j.nima.2013.09.060 | |
| Issue Date | 2014-02-01 | |
| Doc URL | http://hdl.handle.net/2115/54931 | |
| Туре | article (author version) | |
| File Information | Manuscript20130913.pdf | |



| 1 | Energy resolution of pulsed neutron beam provided by the ANNRI beamline | |
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| 4 | K. Kino ^{a,*} , M. Furusaka ^a , F. Hiraga ^a , T. Kamiyama ^a , Y. Kiyanagi ^a , | |
| 5 | K. Furutaka ^b , S. Goko ^{b,1} , K. Y. Hara ^b , H. Harada ^b , M. Harada ^b , K. Hirose ^b , | |
| 6 | T. Kai ^b , A. Kimura ^b , T. Kin ^{b,2} , F. Kitatani ^b , M. Koizumi ^b , F. Maekawa ^b , | |
| 7 | S. Meigo ^b , S. Nakamura ^b , M. Ooi ^b , M. Ohta ^b , M. Oshima ^b , Y. Toh ^b , | |
| 8 | M. Igashira ^c , T. Katabuchi ^c , M. Mizumoto ^c , J. Hori ^d | |
| 9 | | |
| 10 | ^a Graduate School of Engineering, Hokkaido University, | |
| 11 | Kita 13 Nishi 8, Kita-ku, Sapporo 060-8628, Japan | |
| 12 | | |
| 13 | ^b Japan Atomic Energy Agency, | |
| 14 | 2-4 Shirakata Shirane, Tokai, Naka, Ibaraki 319-1195, Japan | |
| 15 | | |
| 16 | °Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology, | |
| 17 | O-okayama, Meguro-ku, Tokyo 152-8550, Japan | |
| 18 | | |
| 19 | dResearch Reactor Institute, Kyoto University, | |
| 20 | 2-1010, Asashiro Nishi, Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan | |
| 21 | | |
| 22 | Keywords: Pulsed neutron beam; Neutron beam line; | |
| 23 | Neutron energy resolution; Neutron capture cross section; | |
| 24 | J-PARC; MLF; JSNS; ANNRI | |
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| 26 | *Corresponding author. Tel.: +81 11 706 6703; fax.: +81 11 706 6703. | |
| 27 | <i>E-mail address</i> : k-kino@eng.hokudai.jp (K. Kino). | |
| 28 | ¹ Present address: Japan Nuclear Energy Safety Organization, 4-1-28 | |
| 29 | Toranomon, Minato-ku, Tokyo 105-0001, Japan | |
| 30 | ² Present address: Department of Advanced Energy Engineering, Kyusyu | |
| 31 | University, Kasuga, Fukuoka 816-8580, Japan | |
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33 Abstract

34 We studied the energy resolution of the pulsed neutron beam of the Accurate Neutron–Nucleus Reaction Measurement Instrument (ANNRI) at 35 the Japan Proton Accelerator Research Complex/Materials and Life Science 36 Experimental Facility (J-PARC/MLF). A simulation in the energy region 37 38 from 0.7 meV to 1 MeV was performed and measurements were made at thermal (0.76-62 meV) and epithermal energies (4.8-410 eV). The neutron 39 energy resolution of ANNRI determined by the time-of-flight technique 40 depends on the time structure of the neutron pulse. We obtained the 41neutron energy resolution as a function of the neutron energy by the 4243simulation in the two operation modes of the neutron source: double- and single-bunch modes. In double-bunch mode, the resolution deteriorates 44above about 10 eV because the time structure of the neutron pulse splits 45into two peaks. The time structures at 13 energy points from measurements 46 in the thermal energy region agree with those of the simulation. In the 47epithermal energy region, the time structures at 17 energy points were 48 obtained from measurements and agree with those of the simulation. The 49FWHM values of the time structures by the simulation and measurements 50were found to be almost consistent. In the single-bunch mode, the energy 51resolution is better than about 1% between 1 meV and 10 keV at a neutron 5253source operation of 17.5 kW. These results confirm the energy resolution of the pulsed neutron beam produced by the ANNRI beamline. 54

56 1. Introduction

In recent years, intense pulsed neutron beams provided by spallation 57neutron sources have been used to study the neutron-nucleus reaction [1,2]. 58Our team has developed a new instrument, the Accurate Neutron-Nucleus 59Reaction Measurement Instrument (ANNRI) [3] at the Japan Spallation 60 Neutron Source (JSNS) [4] in the Japan Proton Accelerator Research 61 Complex/Materials and Life Science Experimental Facility (J-PARC/MLF). 62 63 One of the aims of ANNRI is to provide accurate neutron capture cross-section data for minor actinides and long-lived fission products, which 64 can be experimentally difficult to obtain for some nuclei, for example, when 65 66 the amount of experimental sample is limited due to high radioactivity and/or isotopic contamination. The intense and high-quality pulsed 67 neutron-beam of ANNRI [5] allowed successful measurement of some 68 problematic materials [6-9]. 69

In the measurement of neutron capture cross sections using pulsed 70neutron beams at ANNRI, the neutron energy is calculated from its 71time-of-flight (TOF) between the neutron source and experimental sample. 7273The finite energy resolution due to the time structure of the pulsed neutron beam has to be taken into account in analyzing the experimental data. 74There are two factors that cause time structures for the pulsed neutron 75beam. One is a slowing of neutrons in the moderator of JSNS. Neutrons are 76 generated in a mercury target through the spallation process caused by a 3 77-GeV proton beam. They are then slowed down in a liquid hydrogen 78moderator by collisions with hydrogen. In this slowing down process, a time 79 80 structure arises in the neutron beam from the statistical nature of the 81 collisions. The second factor is the time structure of the incident proton beam. At JSNS the incident proton beam is normally delivered in a 82 double-bunch scheme. The time interval between the two bunches is 599 ns. 83 For materials and life sciences, where cold or thermal neutrons are used, 84 this scheme is not a problem since the time structure due to the 85 86 slowing-down process dominates. However, for epithermal neutrons, which are used for neutron capture cross-section measurements at ANNRI, the 87 double-bunch structure cannot be ignored. 88

In this paper, we present the results of studies of the time structure of the pulsed neutron beam using both simulation and measurements. The simulation covers the entire energy range for neutron capture cross-section

92 measurements. However, the simulation is based on an assumption that 93 JSNS and ANNRI work perfectly as designed. On the other hand, the 94 measurements can provide practical performance data of ANNRI, although 95 the energy range is limited.

96 2. Simulation

97 2.1 Simulation procedure

We performed a simulation of the neutron source using the Monte-Carlo 98 simulation code PHITS [10] to obtain the time structure of the neutron 99 beam. The procedure used in the simulation is very similar to that 100 101 presented in the reference [5]. We applied the nuclear data from the library 102JENDL3 [11] to all the materials in the simulation model apart from hydrogen in the moderator, for which ENDF/B-VI Release3 [12] was used. 103 The simulation model for the neutron source simulates JSNS and includes 104 the mercury target for spallation reaction, moderators, reflectors, and iron 105shields. The parameters used for the simulation are listed in Table 1 and 106 correspond to the operational conditions of 17.5 kW. An event in the 107 simulation is initiated by the injection of a proton into the mercury target. 108 At JSNS, a 3-GeV proton beam bombards the mercury target at a repetition 109 rate of 25 Hz. In this simulation, all protons impinge on the spallation 110 target at the same starting time whereas the actual proton beam has a time 111 112structure. The time structure of the proton beam was taken into account by convolution after the simulation. The convolution procedure is described in 113 Section 2.2. A 100×100 mm² tally, which records information of particles, 114was placed at the moderator surface perpendicular to the ANNRI beamline, 115and neutrons passing through the tally were counted. Neutrons within a 116117very small solid angle region with respect to the ANNRI beamline were 118 considered, in order to obtain the time structure of the neutrons emitted to the sample position of ANNRI. 119

120 [Table1 about here]

121 2.2 Analysis of the time structures obtained by the simulation

In order to represent the time structure as a function of the neutron energy, we fitted time structures in many narrow neutron-energy ranges by a model function. We used the model function proposed by Ikeda and Carpenter [13],

126
$$\psi(v,t) = \int dt' \phi(v,t') [(1-R)\delta(t-t') + R\beta\theta(t-t')\exp(-\beta(t-t'))] \quad (t>0)$$

127
$$= \frac{\alpha}{2} \left\{ (1-R)(\alpha t)^2 e^{-\alpha t} + 2R \frac{\alpha^2 \beta}{(\alpha - \beta)^3} \left[e^{-\beta t} - e^{-\alpha t} (1 + (\alpha - \beta)t + \frac{1}{2}(\alpha - \beta)^2 t^2) \right] \right\}.$$
(1)

128 Here, $\phi(v,t')$ describes the neutron flux for the slowing-down process in an 129 infinite hydrogenous medium and is expressed as follows:

130
$$\phi(v,t) = \frac{\sum_{s} v}{2} (\sum_{s} vt)^2 \exp(-\sum_{s} vt) \quad (t>0),$$

Where Σ_{s} is the neutron macroscopic cross section and v is the velocity of 131132neutrons. The time t was modified as $t-t_0$. The fitting parameters are t_0 , α , β , R, and a scaling factor for eq. (1). Eq. (1) consists of two physical 133134terms. One is the slowing-down term and the other is the storage term. These are 1-R and R in the ratio $(0 \le R \le 1)$ of the total intensity, 135respectively. Fig. 1 shows examples of the fits. The time structures are well 136 fitted by eq. (1) in this simulation, which uses neutrons from the coupled 137moderator of JSNS, although eq. (1) fails to express the time structure in 138 the case of the decoupled moderator used for another beamline at JSNS [14]. 139The fitting parameters t_0 , α , β , and R are plotted as a function of the 140neutron energy in Fig. 2. These data were fitted by polynomial functions in 141order to express these parameters as smooth functions of the neutron 142energy. A two-dimensional plot of the time structure and neutron energy is 143144shown in Fig. 3, showing the relation between the emission time and energy of neutrons at the moderator surface. The origin of the time axis is the 145incident time of the proton beam on the mercury target. 146

- 147 [Fig. 1 about here]
- 148 [Fig. 2 about here]
- 149 [Fig. 3 about here]

The neutron time structure obtained by the simulation was convoluted 150with the time structure of the proton beam. At JSNS, the proton beam 151normally consists of two bunches separated by 599 ns. In this paper, we call 152this proton beam scheme the double bunch. However, depending on the 153JSNS operation program, the proton beam could be a single bunch. Fig. 4 154shows the time structures of the proton beam during the measurements of 155the neutron time structures. The solid and dashed lines represent the single 156and double bunches, respectively. The FWHM value of each bunch is 60 ns. 157Three examples of the convoluted results are shown in Fig. 5. At low 158159neutron energy (Fig. 5a), the time structures of the single and double

160 bunches are almost the same. However, the time structure is different for the double bunch as the neutron energy increases (Fig. 5b and 5c). Fig. 6a 161 and 6b are two-dimensional plots, which show relations between the time 162structure and neutron energy. In the double-bunch mode, the time structure 163splits into two peaks above about 10 eV. This phenomenon reduces the 164165energy resolution. In addition, the time structure is wider compared to that of Fig. 3 at neutron energies higher than about 10 keV because the time 166167 width of the bunch cannot be ignored compared to that of the slowing-down process in the moderator. 168

169 [Fig. 4 about here]

170 [Fig. 5 about here]

171 [Fig. 6 about here]

172 2.3 Simulated neutron energy resolution

We calculated the neutron energy resolution at the sample position of the 173Ge spectrometer at ANNRI. We used the width of the time structure in 174FWHM based on the results described in Section 2.2. In the case where the 175time structure splits into two peaks, we defined the time width as the time 176between the rising edge of the first peak and the falling edge of the second 177peak. Fig. 7 shows the time width as a function of the neutron energy. Above 178about 10 eV, the effect of the double bunch appears as seen in Fig. 6b. For 179the single bunch, the width approaches about 60 ns as the neutron energy 180 increases. This reflects the width of the proton beam bunch. On the other 181hand, for the double bunch, the width approaches about 600 ns, reflecting 182the time distance between the two bunches. In the TOF technique, the 183uncertainty in neutron energy ΔE is calculated from the difference in 184185energies at $t+\Delta t$ and $t-\Delta t$. Here, t is the TOF between the moderator and experimental sample. If Δt is small compared to t, the energy resolution 186

187
$$\frac{\Delta E}{E}$$
 is related to the time resolution $\frac{\Delta t}{t}$ by the following equation:

188
$$\frac{\Delta E}{E} = 2\frac{\Delta t}{t}.$$

For the Ge spectrometer at ANNRI, the TOF distance is 21.5 m. By using the values of the width in Fig. 7 as Δt , we obtained the energy resolution shown in Fig. 8. For the single bunch, the energy resolution is about 1% or less between 1 meV and 10 keV. For the double bunch, the resolution decreases above 10 eV and is 10 times less than that of the single bunchabove about 10 keV.

- 195 [Fig. 7 about here]
- 196 [Fig. 8 about here]
- 197 **3. Measurements**
- 198 *3.1 Thermal neutron*

We measured the time structures of the neutron beam based on the 199 thermal neutron energy. The measurement set up is shown in Fig. 9. We 200placed a mica sample with dimensions of 50×50 mm² and a thickness of 5 201mm in the beamline, 28.5 m from the moderator. Mica is a silicate mineral 202 203 and has a layered crystal structure. We used a mica sample with a layer interval of 10.4 Å. Diffracted neutrons from the sample were detected by a 204helium-3 proportional counter at an angle of 162 degrees with respect to the 205beamline downstream and a distance of 650 mm from the sample. TOF 206spectra of the diffracted neutrons were obtained. The proton beam had a 207repetition rate of 25 Hz and a double-bunch structure. The JSNS power was 208 120 kW. 209

From the Bragg's law, the interval d, scattering angle θ , and neutron wavelength λ are related as follows:

(2)

212 $\lambda = 2d\sin(\theta)$.

If the product of the wavelength of the incident neutron and a positive 213integer value *n* is equal to the wavelength λ in eq. (2), diffraction occurs. 214The diffraction peaks in the TOF spectra reflect the time structure of the 215216 neutron beam. Figs. 10a and 10b show the TOF spectra under two conditions: the disk chopper, which cuts the frame overlap, was not used for 217218Fig. 10a and used for 10b. The arrows in these figures indicate the expected positions of the diffraction peaks. The numbers above some of the arrows 219correspond to the value n. The n=2 and 3 diffraction peaks in Fig. 10a 220221appear in the second frame. The intensities of the diffraction peaks in Fig. 10b are lower than those in Fig. 10a because we had to increase the neutron 222223counter's discriminator threshold due to noise from the disk chopper. The 224spectra with no peaks in these figures, represent the background, measured by setting the angle of the mica sample off the diffraction condition. The 225background was subtracted from the foreground spectra. We obtained 226sufficient statistics to analyze the diffraction peaks with n=2, 3, 4, 5, 6, 8, 9, 22722810, 11, 12, 13, 14, and 18. Fig. 11 shows comparisons of these diffraction

229peaks with the time structures obtained by the simulation. The minimum and maximum neutron energies for the diffractions are 0.76 and 62 meV. 230 respectively. The value indicated in each figure corresponds to the neutron 231energy of the diffraction. The double bunch does not affect the 232233measurements for thermal neutron energies, as shown in Fig. 6. In Fig. 11, 234the intensities for the simulation data are scaled to those of the measurement data. All the figures show good agreement between the 235measurement and simulation in the intensity range from two to three 236 orders of magnitudes. In addition, both the components of eq. (1) show 237agreement between the measurement and simulation. This result indicates 238239that JSNS and the ANNRI beamline work properly for the thermal neutron energies. 240

241 [Fig. 9 about here]

242 [Fig. 10 about here]

243 [Fig. 11 about here]

The measured diffraction peaks were fitted using eq. (1) to get the 244FWHM values of the time structures to allow a comparison with simulation. 245All the parameters for the neutron time structure described in section 2.2 246were free during the fitting process. In the thermal energy region, the time 247structure of the proton beam is negligible. Fig. 12 shows examples of the fits. 248The experimental data are well fitted by eq. (1). Fig. 13 compares the 249250experimentally obtained parameters α , β , and R with those in Fig. 2. Both sets of parameters are in agreement. 251

252 [Fig. 12 about here]

253 [Fig. 13 about here]

254 *3.2 Epithermal neutron*

For epithermal neutrons, we used the resonances of the neutron capture 255reaction for tantalum-181. The measurement setup is shown in Fig. 14. We 256placed a tantalum foil with an area of 100×100 mm² and a thickness of 0.1 257or 0.01 mm in the beamline, 29.54 m from the moderator. Prompt gamma 258259rays were emitted immediately following the neutron capture reaction. The difference between the time of incidence of the proton beam on the mercury 260target of the JSNS and the time that prompt gamma rays are detected 261enables us to measure the neutron TOF. The time structure of the neutron 262beam is convoluted with the TOF spectrum and can be extracted from the 263264measured TOF spectra for neutron capture resonances. We detected the 265prompt gamma rays by a scintillation detector. The detector consists of three sets of plastic scintillator and photomultiplier tube. The scintillators 266 were 703 cm³ in volume in total and were set at about 100 mm from the 267beamline in a direction normal to the beamline. The detection efficiency for 268269gamma rays by a plastic scintillator is generally low. However, a plastic 270scintillator is very insensitive to background neutrons because the cross section of the capture reactions for light nuclei in the plastic scintillator is 271272small. We simulated the neutron background coming from the tantalum foil and found that it was negligible for the analyzed resonances. The threshold 273level for the signal processing of the detector was set to 1 MeV for gamma 274275rays, taking into account the need for sufficient statistics and background rejection. 276

277 [Fig. 14 about here]

We took two data sets with two tantalum foils, one thick (0.1 mm) and one 278thin (0.01 mm). The data for the thick foil is for the higher energy 279resonances, whose cross section is small. This data was taken in the 280single-bunch operation mode of JSNS. The data for the thin foil is for the 281low energy resonances. For the low energy resonances, the TOF spectrum 282with the thick tantalum foil saturates at the peak of the resonance due to its 283large cross section. The operation of JSNS was the double bunch for the 284data with the thin foil. 285

The measured TOF spectrum with the thick foil overlapped the TOF spectrum of the evaluated cross section at a temperature of 300 K in the nuclear data library JENDL-3.3 [11] in Fig. 15. As seen in this figure, we observed resonance peaks of Ta-181 at neutron energies from 4.3 eV up to about 400 eV.

[Fig. 15 about here]

We extracted the time structures of neutron pulses from the measured 292TOF spectra using the following procedure. First, we obtained the neutron 293pulses by a convolution of the time structure modeled by eq. (1) and the time 294295structure of the proton beam. Second, the neutron pulses were convoluted with the TOF spectra of neutron capture resonances, which are expressed 296 by the single-level Breit-Wigner equation. The resonance parameters used 297in the single-level Breit-Wigner equation were those in JENDL-3.3. Here, 298we took into account the Doppler effect on resonances using the technique of 299300 effective temperature [15]. Finally, we fitted the TOF spectra obtained as

explained above to the measured spectra. All of the parameters for the 301 302 neutron time structure described in section 2.2 were free during the fitting process. Among the many resonances, we chose isolated ones, namely the 303 304 resonance with the least overlap with neighboring resonances. Fig. 16 305 shows examples of fits for the resonances of energies 4.28, 20.29, and 208.48 306 eV. In Figs. 16a and 16b, the spectra have symmetrical shapes because the time structure of the neutron pulses is narrow compared to the resonance 307 308 width, and the Doppler broadening affects the TOF spectra symmetrically for Ta-181 resonances at room temperature. On the other hand, the 309 spectrum in Fig. 16c shows an asymmetrical shape, which reflects the time 310 311 structure of the neutron pulses. Fig. 16 also shows the resonances calculated using the parameters in Fig. 2. The resonance shapes were well 312reproduced by the simulation. Fig. 17 compares the experimentally 313 obtained parameters α , β , and R with those in Fig. 2. Both sets of 314parameters are in agreement. 315

The FWHM values of the neutron pulses for single-bunch mode were obtained. We used the TOF spectrum with the tantalum foil of thickness of 0.1 mm for all the resonances except the 4.28 and 10.36 eV resonances, for which the double-bunch structure of the proton beam was taken into account for fitting.

- 321 [Fig. 16 about here]
- 322 [Fig. 17 about here]

323 4. Comparison of the simulation and measurements

324 We compared the time structures of the neutron pulses between the simulation and measurements. Fig. 18 shows the FWHM values 325 326 corresponding to those of the single bunch. The error bar for each point, which originates from the statistical uncertainty of the measurement 327 spectrum, is within the size of the marker. The trend and absolute values of 328 329 the measurement results are almost reproduced by the simulation. This result implies that JSNS and the ANNRI beamline work properly and the 330 331 simulation is reliable for deducing the FWHM values in the energy regions where the measurements data were not obtained. 332

333 [Fig. 18 about here]

334 5. Conclusions

We have performed a simulation and performed measurements of the time structure of the neutron pulses at the ANNRI beamline, to obtain accurate data of the neutron-capture cross-sections for minor actinides andlong-lived fission products.

The simulation, which models the neutron source precisely, predicted the 339 time structure for neutron energies between 0.7 meV and 1 MeV. From this 340 we obtained the energy resolution that is determined by the TOF technique. 341342For the double-bunch mode, the energy resolution was found to deteriorate above about 10 eV, demonstrating that we need a special method to analyze 343 experimental data at high energy resolution. We made measurements in the 344thermal and epithermal energy regions with different methods. In the 345thermal energy region, time structures at 13 energy points were measured 346 347 using diffraction by a mica sample. The shapes of the time structures were in agreement with those of the simulation predictions. In the epithermal 348 energy region, we obtained the TOF spectra for neutron capture resonances 349 by tantalum-181 nuclei. From these spectra, we extracted time structures at 350 17 energy points. 351

The FWHM values of the time structures of the neutron pulses for both the simulation and measurement data were in good agreement with regard to the trend and the absolute value. This result shows that the neutron source and ANNRI beamline are working properly, and the reliability of the simulation is also confirmed. In single-bunch mode, we found that the energy resolution was better than about 1% in the energy region from 1 meV to 10 keV.

Currently, the power of JSNS is increasing and will reach 1 MW in the near future. The properties of the proton beam, such as the time width of the beam bunch and the spatial distribution on the mercury target, may change with the increase in power. Therefore, it is important to periodically check the time structure of the neutron pulses by simulation and measurement.

365 Acknowledgments

Present study is the result of "Study on nuclear data by using a high intensity pulsed neutron source for advanced nuclear system" entrusted to Hokkaido University by the Ministry of Education, Culture, Sports, Science, and Technology of Japan (MEXT). This work was supported by JSPS KAKENHI Grant Number 22226016.

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- 393
- 394

395 Figure captions

396

397 Figure 1

Examples of fits of eq. (1) to the time structures of neutron pulses obtained by the simulation. The solid lines represent eq. (1). The dashed and dotted lines correspond to the slowing-down and storage terms in eq. (1), respectively. The values indicated in the figures are neutron energies.

402

403 Figure 2

404 Fit parameters t_0 , α , β , and R. The solid lines are polynomial functions, 405 which were fitted to the data points.

406

407 **Figure 3**

Two-dimensional plot of the time and energy of neutrons at the moderator surface. The time structure of the proton beam is not convoluted. The intensity is normalized at the pulse peak. This plot is drawn at 1/100th of the intensity of the peak.

412

413 **Figure 4**

Time structures of the proton beam for the JSNS operation conditions when the measurements were performed. The solid and dashed lines represent the single-bunch and double-bunch modes, respectively. The FWHM value of each bunch is 60 ns.

418

419 **Figure 5**

Examples of convolution of the time structure originating from the
slowing-down process in the moderator with the proton-beam time structure.
The neutron energies of figs a, b, and c are 1.02 meV, 40.7 eV, and 1.02 keV,
respectively. The solid and dotted lines are the results for the double- and
single-bunch modes, respectively.

425

426 **Figure 6**

Two-dimensional plots of the time and energy of neutrons at the moderator
surface. The time structure of the proton beam is taken into account.
Figures a and b are the results for the single- and double-bunch modes,
respectively.

432 Figure 7

The FWHM values for the time structure by the simulation as a function of
neutron energy. The solid and dashed lines represent the single- and
double-bunch modes, respectively.

436

437 **Figure 8**

438 Neutron energy resolution at 21.5 m from the moderator based on the
439 simulation. The solid and dashed lines represent the single- and
440 double-bunch modes, respectively.

441

442 **Figure 9**

443 Measurement setup for the time structure of neutron pulses at thermal444 neutron energies.

445

446 **Figure 10**

TOF spectra obtained by the diffraction method. The disk chopper was not operated for figure a and was operated for figure b. The spectra, with no peaks are the background data, which were taken by setting the angle of the mica sample off the diffraction condition. The arrows in these figures indicate the expected positions of the diffraction peaks. The numbers above some of the arrows correspond to the parameter n, as explained in the text.

453

454 **Figure 11**

455 Comparison of measured diffractions (data points) with the time structures 456 obtained by the simulation (solid lines). The value indicated in each figure 457 corresponds to the neutron energy of the diffraction. The dashed and dotted 458 lines are the slowing-down and storage terms in eq. (1), respectively.

459

460 Figure 12

461 Examples of fits of the measured data with eq. (1). The solid lines represent
462 the fitting results. The dashed and dotted lines are the slowing-down and
463 storage terms in eq. (1), respectively.

- 464
- 465
- 466
- 467

468 **Figure 13**

469 Comparison of parameters α , β , and R in the thermal neutron region. 470 The lines and data points are the simulation and measurement data, 471 respectively.

472

473 **Figure 14**

474 Measurement setup for the time structure of neutron pulses in the 475 epithermal neutron energy region.

476

477 Figure 15

478 Measured TOF spectrum with the thick tantalum foil and the cross section
479 of the neutron capture reaction based on the nuclear data library
480 JENDL-3.3 at a temperature of 300 K.

481

482 Figure 16

Examples of fits of the measured TOF spectra in the epithermal neutron
energy region. Figures a, b, and c show the resonances with energies of 4.28,
20.29, and 208.48 eV, respectively. Resonance curves are also evident, which
are calculated using the parameters obtained by the simulation.

487

488 **Figure 17**

489 Comparison of parameters α , β , and R in the epithermal neutron region.

490 The lines and data points are the simulation and measurement data,

- 491 respectively.
- 492

493 **Figure 18**

- 494 Comparison between the simulation and measurements of the FWHM
- 495 values of the time structures of neutron pulses for single-pulse mode.
- 496

Table 1

| Proton beam | Energy | 3 GeV |
|---------------------|------------------|--|
| | Spatial shape | Rectangle (uniform distribution) |
| | | -25.9 – $+25.9$ mm (horizontal) |
| | | -12.6 - +12.6 mm (vertical) |
| $Liq.H_2$ moderator | Temperature | 19.7 K |
| | Density | $4.2655\!	imes\!10^{22}$ atoms/cm 3 |
| | Ortho-Para ratio | Para 100% |
| Cooling water | Reflector | Light water |
| | Mercury target | Light water |

Parameters used in the simulation of the neutron source.















FWHM (µs)







TOF (µs)

TOF (µs)









Neutron Energy (eV)







