Recent Progress on DNA ToF Backscattering Spectrometer in MLF, J-PARC

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Abstract. DNA is a time-of-flight backscattering neutron spectrometer installed at the Materials and Life Science Experimental Facility in J-PARC. After opening user program, we have been developing and upgrading DNA. The energy resolution is improved and achieved the designed value of 1.4 μeV by renewing the disks of the pulse shaping chopper that can rotate at max 300 Hz instead of 225 Hz of the previous disks. The installation of Si311 analyser mirrors has been almost accomplished, which extends momentum transfer region up to 3.7 Å^{-1}. We demonstrate the effect of this extension by measuring and analysing methyl rotation dynamics in solid toluene at 10 K and 40 K. We also report variety of sample environment available at DNA and their remote handling.

1 Introduction

DNA is a time-of-flight (tof) backscattering spectrometer [1] installed at BL02 neutron port seeing a coupled moderator with the largest intensity in a cold neutron wavelength among three types moderators [2] of the Materials and Life Science Experimental Facility (MLF) in the Japan Proton Accelerator Research Complex (J-PARC). Apart from the name of DNA biomolecular dynamics spectrometer, it aims at revealing pico to nano second dynamics in a wide field of basic sciences and industrial applications including soft matter, bio-molecules, chemical molecules, battery, catalyst, and magnetism. Since it has been opened to users from March 2012, the fields of publications are distributed as 34 % liquid, 18 % polymer, 16 % biomatter, 11 % crystal chemistry, 10 % magnetism, 3 % ionic protonic conduction, 3 % hard matter (lattice dynamics) together with 5 % instrumentation and sample environment.

DNA has an advantage of flexibility of various energy resolution by utilizing a pulse-shaping chopper, PSC, which has counter-rotating double disks with four slits (three 3-cm slits and one 1-cm slit) [1]. The PSC is located at 7.75 m downstream of the coupled moderator and plays a role of a pseudo pulsed neutron source with the multiple slit-openings and the flexible open-timing, which gives us an interesting and useful function of scanning a wide energy-transfer region with keeping good resolution. [1] The shift of the slit opening phases enables us to scan a wide energy transfer, *E*, range from -600 to 1500 μeV for Si111 analyzers even with the highest resolution.

DNA has another advantage of extremely good signal-to-noise ratio of more than 10^5 which is attributed by a curved neutron guide cutting high-energy neutron and a back-coating of Gd metal or Gd_2O_3 of the analyzer mirrors.

In this paper, we will report recent progress on DNA which has been achieved after the previous report of the instrumentation [1]. We will introduce an improvement of the energy resolution by re-fabricating the disks for the PSC in §2, and the extension of Si311 analyzer mirrors in §3. In §4, variety of sample environment available in DNA will be introduced along several types of experiment. In §5, near-future plan on the instrumentation for DNA will be explained. All contents of the paper will be summarized in §6.

2 Improvement of Energy Resolution

The new disks for the PSC, which are thicker by 1 mm than the old disks, were re-fabricated to improve mechanical strength, because a large vibration of the old disks limits the maximum rotation speed to 225 Hz instead of the designed 300 Hz. The energy resolution is determined by (1) mirror factor originating from the Bragg reflection angle and the mosaicity of the analyzer mirrors and (2) tof factor defined by time width of the pseudo pulsed neutron source defined by the PSC rotating speed. The contributions depend on neutrons with the final energy determined by Bragg law. The first factor is constant by geometry of Bragg angle of 87.5° and the mosaicity of the mirror material while the second factor becomes smaller with faster rotation of PSC.

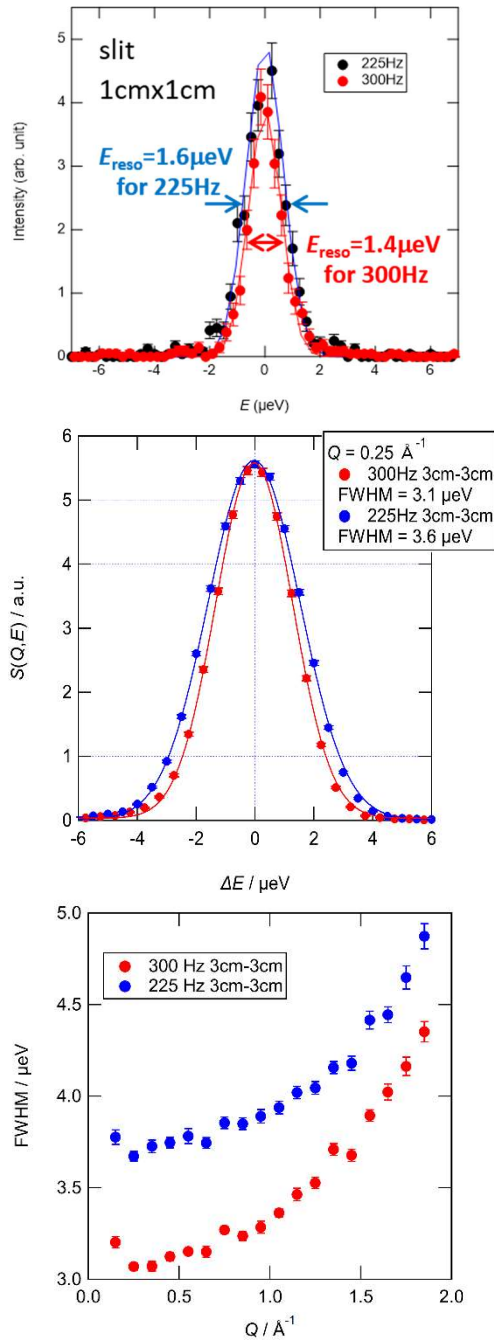* Corresponding author: kawakita.yukinobu@jaea.go.jp

Fig. 1. The energy resolutions of Si111 analyzer mirrors under the highest-resolution condition of the sample size of 1 cm (D) × 1 cm (H) with the slit size of 1 cm × 1 cm (top) and the conventional condition of the sample size of 1.4 cm (D) × 4 cm (H) with the slit size of 3 cm × 3 cm (middle). The *Q*-dependences of the energy resolutions for these conditions are compared in the bottom.

DNA is designed to have both terms balanced at 300 Hz rotation. The 300 Hz PSC operation improved the best performance of the energy resolution for Si111 analyzer mirrors from 1.6 μeV to 1.4 μeV for 1 cm (D) × 1 cm (H) sample size with 1 cm × 1 cm slits, and the conventional resolution from 3.6 μeV to 3.1 μeV for 1.4 cm (D) × 4 cm (H) of sample size with 3 cm × 3 cm slits as shown in Fig. 1. The figures of the quasi-elastic neutron scatterings (QENS) spectra were plotted with an arbitrary vertical scale because of easy comparison for their energy
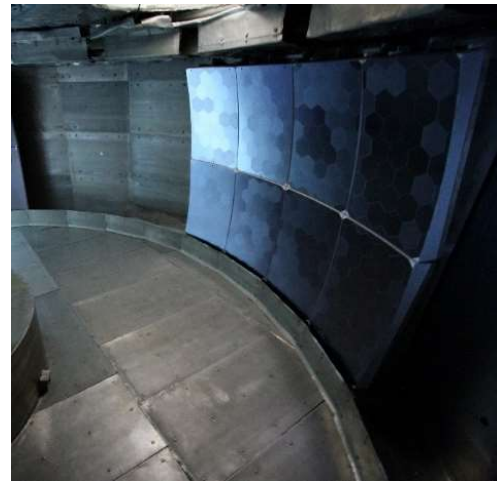

Fig. 2. The photo of the Si311 analyzer mirrors.
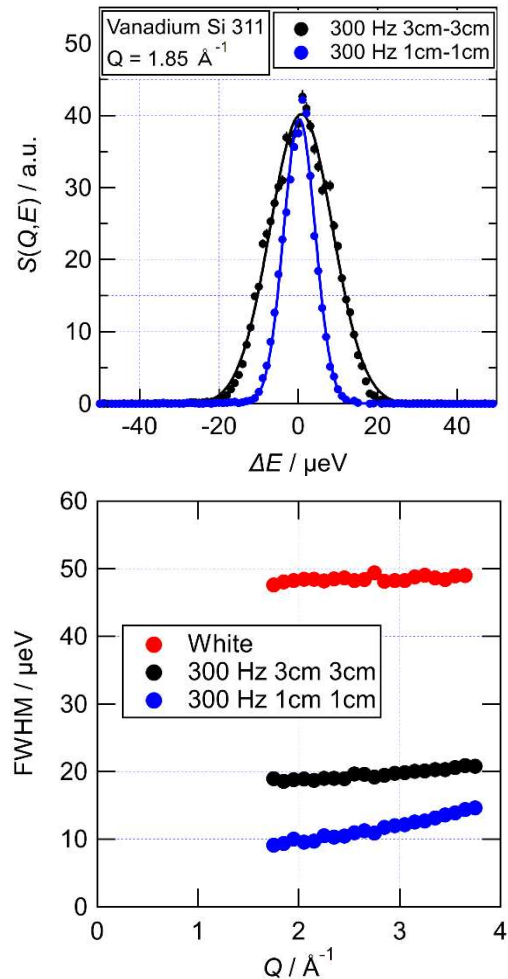

Fig. 3. The energy resolutions of Si311 analyzer mirrors under the conventional condition of the sample size of 1.4 cm (D) × 4 cm (H) with the slit size of 3 cm × 3 cm and 1 cm × 1 cm (Top). The *Q*-dependences of the energy resolutions for the Si311 analyzer is shown in the bottom. "White" (red) corresponds to the condition without operating PSC.

resolutions. The ratio of the integration of the spectrum at 300 Hz to that at 225 Hz is around 52 %, which almost

equals to the square of 225/300 derived from the beam slice by the counter-rotating double disks.

As an example that the finest energy resolution would be useful, hyperfine splitting of Nd nuclei in Nd compounds was studied by utilizing DNA[3]. In their study, PSC condition of 225 Hz but small sample size leaded to a relatively fine resolution of around 2 μeV. They were able to distinguish hyperfine contribution aside the elastic peak. To utilize the finest resolution with 300 Hz rotation would give much clearer separation between them. Such very low energy lying excitation could be a target of DNA.

3 Extension of Analyzer Mirrors

In addition of the Si111 analyzer mirrors covering horizontal scattering angles from -30° to 150° (vertical ones from -14° to 21°), Si311 analyzer mirrors covering horizontally from -150° to -55.5° have been installed year by year as shown in Fig. 2. While the Si111 covers momentum transfer, Q, range is from 0.08 to 1.98 Å^{-1}, Si311 extends Q-range to 3.7 Å^{-1}. The energy resolutions of Si311 under various conditions were examined by measuring a vanadium standard sample, as shown in Fig. 3. The highest resolution of 10 μeV is achieved by 1 cm × 1 cm slits with 300 Hz PSC operation. The intensity is two order weaker than that for Si111 with 300 Hz and 3 cm × 3 cm slits. The useful energy resolution of 19 μeV is obtained by 300 Hz with 3 cm × 3 cm slits. Without the PSC operation, it is 50 μeV with a reasonable intensity. The Q-E ranges measurable by Si111 and Si311 are individually shown in Fig. 4. It is noted that simultaneous measurement by using Si111 and Si311 analyzers is impossible because the energy band to be introduced to the sample through PSC and the frame choppers is different between Si111 and Si311 analyzers. Although it is not shown in Fig. 4, Si333, the 3rd harmonics of Si111, has a very weak intensity and limited energy band due to
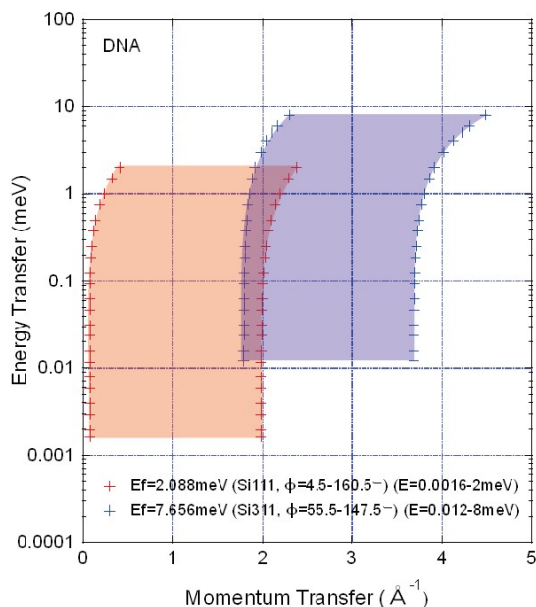
the curved mirror guide cutting high energy neutrons and it is available only for alignment check of single crystal whose QENS or inelastic neutron scatterings (INS) will be measured.

To demonstrate the performance of the extending Q-E region, we measured QENS spectra for methyl rotation in solid toluene at 10 K and 40 K. Fig. 5 shows some combinations of Si111 and Si311 analyzers. At 10 K, tunneling modes of methyl rotation [4] are observed at around ±30 μeV as shown in Fig. 5(a). In such case, the combination of both the high resolution modes of 3.6μeV for Si111 with 225 Hz and 3 cm × 3 cm slits and 10 μeV for Si311 with 300 Hz and 1 cm × 1 cm slits are useful. The measurement time may be a few ten minutes for Si111 and several hours for Si311. Figures 5 (b) and (c) show the spectra at 40 K where only QENS signal is observed with the combinations of the high flux mode of Si111 without PSC (13 μeV in resolution) and (b) the high resolution mode of Si311 with 300 Hz and 3 cm × 3 cm slits (19 μeV in resolution) and (c) the high flux mode of Si311 without PSC. The latter exhibits the widest energy region accessible by DNA.

The QENS profile at 40 K was fitted by a sum of a delta function ($\delta(E)$) representing the elastic component and a Lorentz function ($L(\Gamma,E)$) representing the QENS component convoluted with a resolution function ($R(Q,E)$), with constant background (BG) as follows,

$$S(Q,E) = [A_E\delta(E) + A_L(\Gamma,E)L(\Gamma,E)] \otimes R(Q,E) + BG \tag{1}$$

where the A_E and A_L are coefficients of the delta function and the Lorentz function, respectively. The Γ is a half width at half maximum of the Lorentz function. Figures 6 (a) and (b) show a typical fitting profile at $Q = 2.35$ Å^{-1} and the Q dependence of the Γ, respectively. The elastic incoherent scattering factor (EISF) was calculated as follows.

$$EISF = \frac{A_E}{A_E + A_L} \tag{2}$$

In Fig. 6(c), the calculated EISF is plotted together with a theoretical curve assuming methyl rotation,

$$EISF = \frac{5}{8} + \frac{3}{8}\left[\frac{1+2j_0(Qr)}{3}\right] \tag{3}$$

where the first term was due to the benzene ring and the second term represents a three site jump of the methyl group[5]. The j_0 and r are the zeroth-order spherical Bessel function and H-H distance (=1.78 Å) in the methyl group. One can understand the effectiveness of QENS measurement in a wider Q region.

Difference in a jump length of the jump diffusion model can be more clearly seen in a high Q region, which means that this combination method has an ability to distinguish an ionic conduction path among possible paths determined by powder diffraction. Coherent QENS measurements can be also a target for the combination method of Si111 and Si311 analyzers.
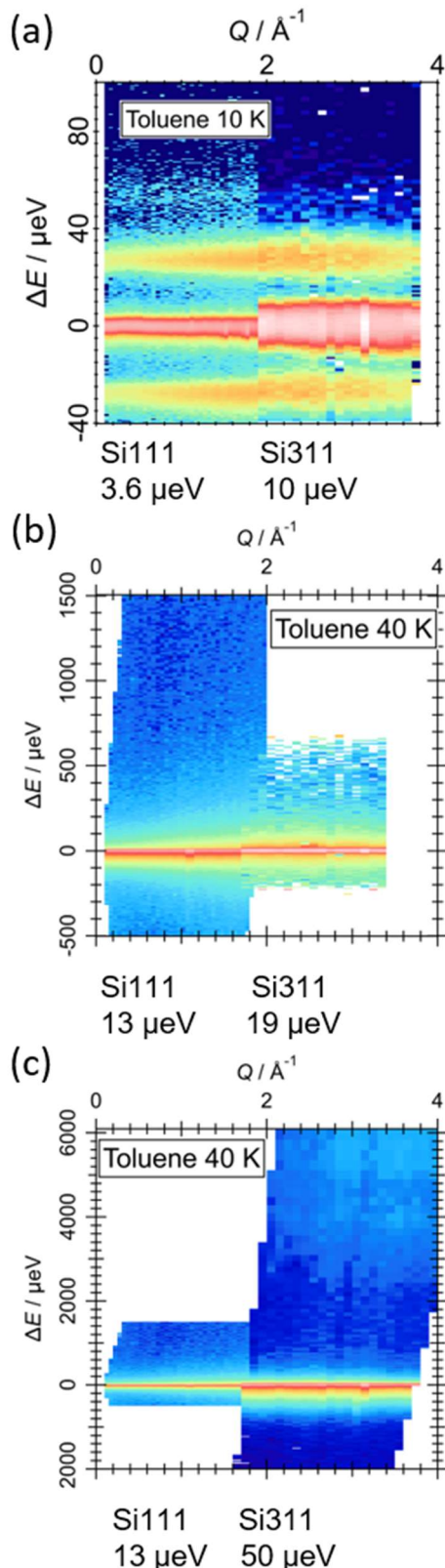

Fig. 4. The wide Q-E region measurable by DNA. The areas for Si111 and Si311 are hatched by orange and purple.

Fig. 5. The QENS spectra for solid toluene at 10 K and 40 K with various energy resolutions obtained with Si111 and Si311. (a) The combination of 3.6 μeV for Si111 (225 Hz, 3 cm × 3 cm slits) and 10 μeV for Si311 (300 Hz, 1 cm × 1 cm slits), (b) the combination of 13 μeV for Si111 (without PSC) and 19 μeV for Si311 (300 Hz, 3 cm × 3 cm slits), and (c) the combination of 13 μeV for Si111 (without PSC) and 50 μeV for Si311 (without PSC)
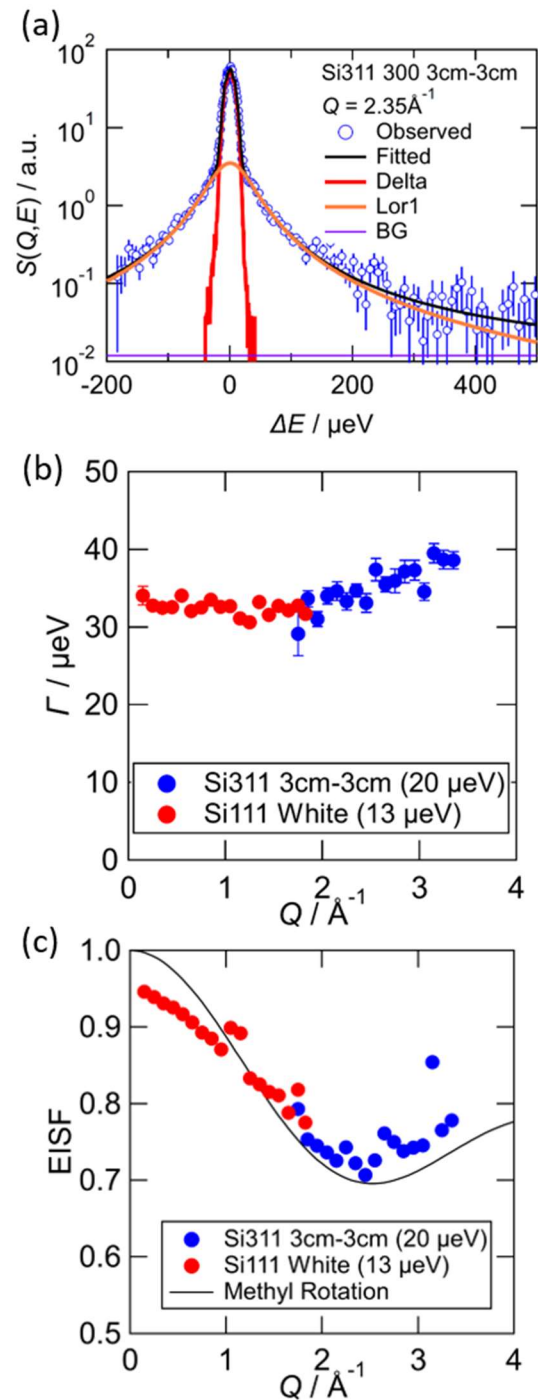

Fig. 6. QENS analysis of toluene at 40K. (a) A curve fitting with a delta plus a Lorentian functions together with a constant background which are convoluted by instrument resolution function. (b) The Q dependence of the Lorentzian width. (c) The elastic incoherent structure factor together with a theoretical curve assuming methyl rotation.

One of coauthors (MM) studied ionic diffusion in Li based solid electrolyte [6]. In their study, single crystal was mounted to access the (hhl) horizontal scattering plane and rotated to make scattering trajectories nearly parallel to the three principal axes [110], [111] and [001]. The Q-dependence of the energy width of the QENS signal is different along three principal axes, which makes us determine ionic diffusion path. Since

the difference appears more clearly in a high Q region, Si311 would be powerful to see it.

4 Variety of Sample Environment

In DNA, many kinds of sample environment (SE) are available according to wide variety of target sciences. In this section, such SEs are introduced and a remote control and automation are focused.
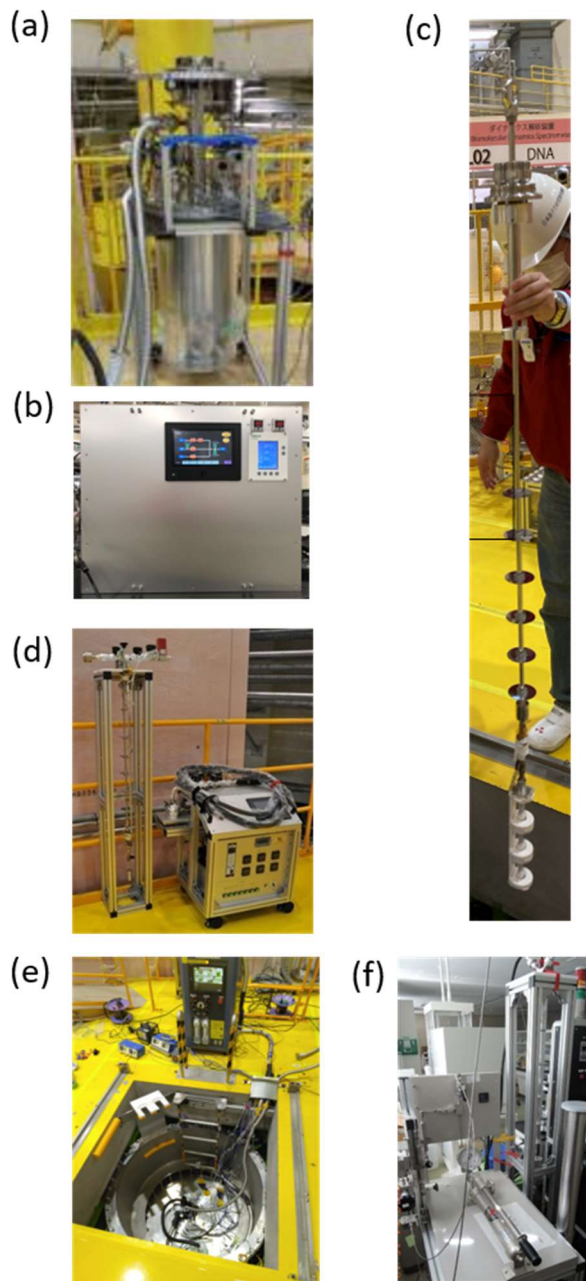

Fig. 7. (a) Standard cryo-furnace at DNA (5 K < T < 700 K), (b)gas handling system DICE, (c)sample stick with automatic sample positioning function for three samples, PEACE. (d)humidity control system, (e)new humidity control system with gas mixing function (common SE), (f) hydraulic press system (0.1 < P < 145 MPa).

4.1 Standard SEs

A standard SE is a cryo-furnace available in a temperature range from 5 K to 700 K (Fig. 7(a)). The cryo-furnace has two types of vacuum chamber, namely an outer vacuum chamber (OVC) which stores a cold head of the closed cycle refrigerator (CCR) and an inner vacuum chamber (IVC) which is a space for installation of a sample stick with an internal heater. The CCR cools IVC and then the sample stick through dilute He gas (approximately 8×10^3 pascals) inserted to IVC, while heating of the sample stick is carried out by the internal heater with thermal isolation from the cool IVC by evacuation. An automated gas-handling system named as DICE [7] developed by the DNA team (Fig. 7(b)) is always auxiliary used together with the cryo-furnace. As well as several standard sample sticks, a stick with sample changer function with positioning three samples vertically arranged (named as PEACE) is frequently utilized in a temperature range below 340 K [7] (Fig. 7(c)).

4.2 SEs for Soft-matter, Bio-molecule and Polymer Sciences

Humidity is an important control parameter for polymer science. DNA has its own humidity control system as shown in Fig. 7(d) and a corresponding sample stick to be installed into the cryo-furnace. Based on this technique a new one which can provide vapor gas mixed H_2O and D_2O at a favorite concentration flexibly has been developed as a common SE device at MLF (Fig. 7(e)). This new machine is also available.

A special sample stick with pressure generator which hydraulic pressure up to 145 MPa [8] can be applied is available at the room temperature (Fig. 7(f)).

4.3 SEs for Energy Science and Solid-State Physics

Two kinds of special purpose sample stick were developed. One can apply electrical current up to 2 A to the sample (Fig. 8(a)), while the other can apply high voltage up to 1.5 kV to the sample (Fig. 8(b)). Measurements under current or voltage may accelerate science of battery electrolyte and multiferroics, pharmaceutical application such as drug delivery. An air-tight sample cell with electrodes is developed for in situ current experiments as shown in Fig. 8(c).

For solid state physics, extremely low temperature is required, which is realized by the combination of liquid He type 2 K cryostat (Blue Cryo) (Fig. 8(d)) and a dilution option (common SE in MLF) (Fig. 8(e)). The Blue Cryo itself is available in the temperature range from 300 K to 1.5 K and with the dilution option 60 mK is achieved.

4.4 Sample cells

Including the sample cell for current and voltage application described above, we developed several sample cells such as standard aluminum cells for cooling and heating, quartz glass cell [9], single crystal sapphire

cell, molybdenum cell for high temperature. We also investigated corrosion of aluminum cell by solution [10,11] and proposed surface treatment of aluminum cell adequate for some cases [12]. The details are shown in the references.
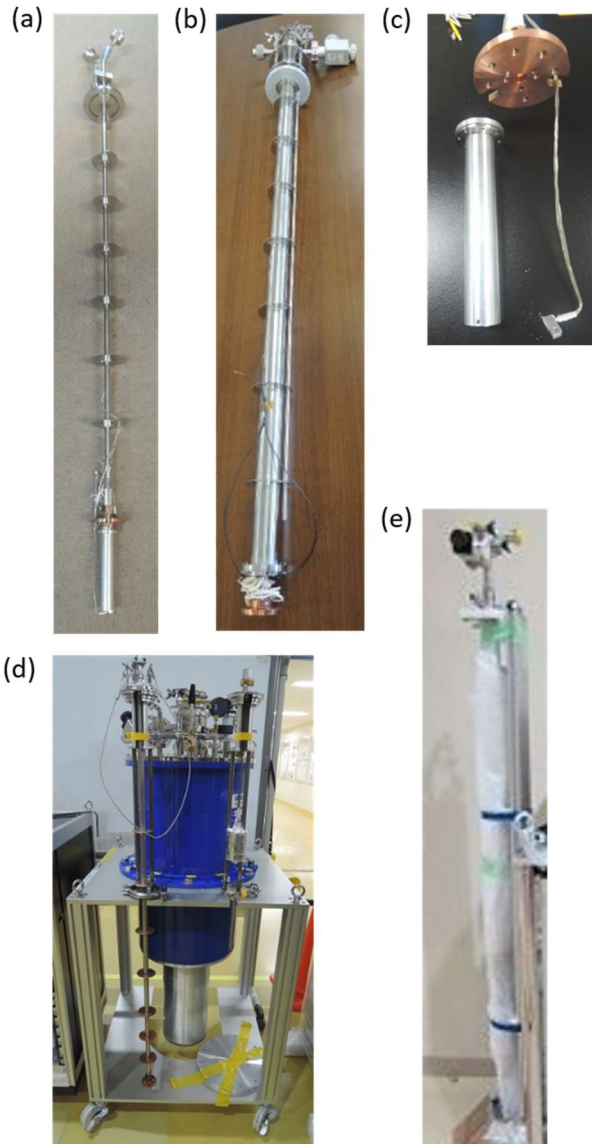

Fig. 8. (a)Sample stick for current application (Max 2 A, T < 500 K), (b)sample stick for voltage application (Max 1.5 kV, T < 500 K), (c)air-tight sample cell with electrodes, (d) liquid He type 2K cryostat (Blue Cryo) (1.5 K < T < 300K), (e) dilution insert (60 mK with the Blue Cryo)

4.5 Automation and Remote Control of SEs

Many of SEs can be controlled from the software platform called as IROHA2 [13] which unifies the data acquisition system (DAQ) and device control. For almost every SE, middlewares controlling its operation parameter such as current, voltage, vacuum level, position encoding etc. and monitoring its environment values such as temperature, pressure, humidity etc. are developed to work on the
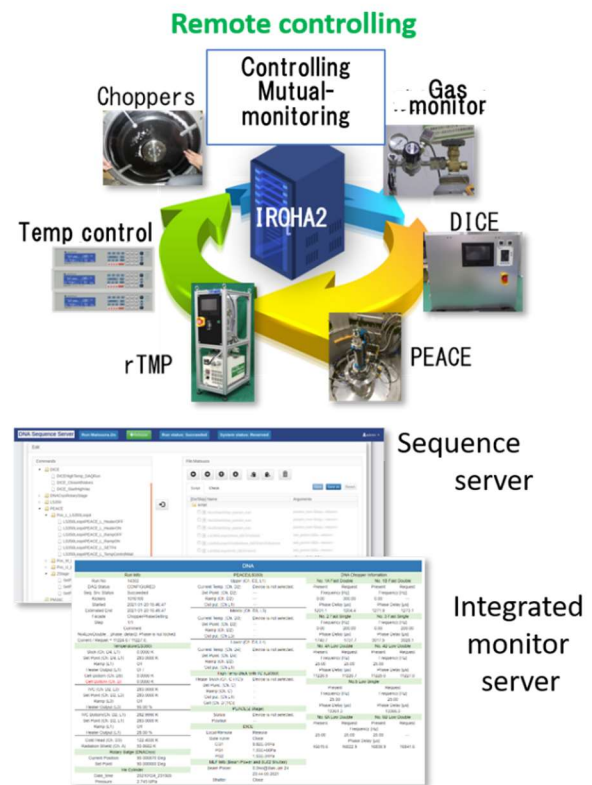

Fig. 9. Remote controlling way at DNA on the software framework IROHA2[13]. From the sequence server, command lists according to timeline of experiment can be conducted remotely. All status of devices is watched by the integrated monitor system.

IROHA2 platform. In the sequencer window, successive commands are arranged to make timelines to be done which involves also ques to start DAQ and end it and therefore we can control all devices and measurement sequence automatically. IROHA2 has also the integrated monitor system showing all status of the instrument which we can watch from outside of the facility. The status of the remote controlling at DNA is schematically shown in Fig. 9. We can watch a residual gas pressure of the gas cylinder such as He gas frequently used for inert gas for IVC of the cryo-furnace and operate a turbo molecular pomp (rTMP) remotely, as well as PEACE, DICE, temperatures of sample sticks, and choppers including PSC.

5 Near Future Plans of DNA

In the summer 2022, we plan to replace the converging neutron guide supermirrors in the vacuum chamber of DNA to new mirrors to improve neutron reflectivity. The guide jacket will be also renewed to non-magnetic materials. Then a small superconductor magnet with max 3.5 Tesla fitted to the small 400A flange of DNA will be commissioned. A high temperature furnace is also planned to be introduced. Two types of oscillatory radial collimator with different collimation degree are fabricated and under commissioning. These will be utilized to the cryo-furnace, the Blue Cryo, the high temperature furnace to eliminate unfavourable scattering from the wall of IVC

and OVC, a container for single crystal with relatively large diameter, thermal shields and so on.

6 Summary

In this paper, we report recent progress including near-future plans on DNA. The improvement of the energy resolution was achieved by re-fabricating the disks for the pulse shaping chopper. Si311 analyzer mirrors extend accessible Q region with high energy resolution (~10 μeV). Variety of sample environments are available to user experiments. The auto and/or remote operations have also been developed.

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