Synchronized UCN storage for superthermal SD$_2$ converter

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SUMMARY OF THE PROPOSAL

We propose to experimentally verify a principle of a substantially new type of compact source for Ultra Cold Neutrons (UCN) in support of designing a practical source dedicated to researches on fundamental particle physics in the 21st century such as the electro-weak interaction, the new models beyond the standard model, the gravity, and so on with improved sensitivity.

The novelty of this UCN source is to efficiently accumulate UCN emitted in a pulsed scheme from a surface of a superthermal converter, solid deuterium (SD$_2$) by using a large kick-out velocity (~5 m/s) of UCN from the SD$_2$ surface and a moving membrane synchronized with pulsed proton beam. This will enable us to make a much more compact UCN source with a performance higher than others ever constructed.

We perform experimental verification of the proposed concept by making use of a prototype assembly consisting of the SD$_2$ converter, a moving membrane, a compact UCN storage vessel and a UCN detector with pulsed proton beam.

As a promising pre-moderator, solid mesitylene (C$_6$H$_3$(CH$_3$)$_3$) operated at low temperatures ($\leq$ 20 K) is examined on the suitability for a pre-moderator satisfying a time structure condition required for the present method of synchronized UCN storage. This test will be done by observing the time dependence of the intensity and the energy spectra of cold neutrons from solid mesitylene with the TOF method. For this purpose, we will observe the time dependences of energy spectra for produced cold neutrons emerging out of the mesitylene pre-moderator by a TOF method.
I. INTRODUCTION

Neutrons with kinetic energies of hundreds of neV are called ultra cold neutrons (UCN). Since their energies are extremely low enough to be trapped in material bottles, they have been used as a probe for investigation of a variety of fundamental researches such as measurement of the neutron lifetime, neutron electric dipole moment, and so on [1]. In this section, we will describe the aim of the present proposal after shortly giving the background surrounding the present status of the UCN studies.

A. Brief History of UCN Production

In 1946, Fermi and Zinn experimentally demonstrated the total reflection of cold neutrons at low grazing angles by the effective potential at material surfaces, where the neutron wave length is enough long compared with inter-atomic spacing [2]. Zel'dovich suggested that this effect could be used to bottle neutrons with kinetic energies below the effective potential of the bottle walls [3] after encouraged by the Fermi's result. Following this suggestion the existence of these neutrons was first demonstrated independently by Luschikov et al. [4], and Steyerl [5]. Since then, UCN densities at reactor sources have gradually increased with reactor power and with improved techniques for extracting the UCN flux. As a result, UCN with the highest density of about 50 UCN/cm$^3$ obtained at the ILL reactor with the rated power of 58 MW in Grenoble have been utilized over these 15 years [6].

In recent years, cryogenic UCN sources using spallation neutrons produced by high energy proton beam opened an experimental chance in accelerator facilities to study the fundamental neutron physics with UCN whose density could amount to the value much higher than the reactor sources.

In fact, very recently, the spallation neutron facility at Los Alamos (LANSCE) succeeded in producing UCN density ($\rho_u = 130$ UCN/cm$^3$), the highest in the world by using a superthermal SD$_2$ converter stored in a production vessel with reflective walls and a spallation neutron source driven by the 800-MeV proton beam whose maximum power is 80 kW [7].

B. General view of production method of UCN

The methods to generate the UCN are, in general, based on either the Doppler shift method or the converter method. Though the source of the former type has provided high UCN densities at the ILL reactor, Grenoble, the latter one has nowadays become more prevalent as a world standard because of more promising potentiality. As far as the converters now practically working or close to launch are concerned, only a superfluid He (He II) [8–14] and a solid deuterium (SD$_2$) [15–17,7] converters are nowadays available.

The concept of the He II is that phonon created in the liquid by a coherent neutron scattering can be used to energetically down-scattered cold neutrons to the UCN regime, while up-scattering can be completely suppressed by maintaining the superfluid at sufficiently low temperature. Thus, neutrons are cooled down much lower temperature than that of the converter itself. In this respect, it is reasonable to give a name a superthermal method to this method.
The density of UCN accumulated in the He II is determined only by the wall absorption and the intrinsic neutron lifetime. Golub et al. [8] showed that UCN were accumulated in the He with an amount comparable to the theoretical estimation. However, since the UCN density estimation was done by an indirect method to measure the up-scattered cold neutrons, further direct verification is required.

Though the He II seems to be attractive, the fact that potential energy of the He II is far smaller than that of the SD$_2$ may be a serious drawback of the He II converter because of difficulty in extracting UCN out of the He II converter. In fact, it was reported that the extracted UCN density was only $\sim 1/50$ of those in the He II [18]. As long as UCN physics is performed inside of the He II container, the experiment will be going successfully. However, if the UCN experiment is performed in a separate vessel after extracting them from the He II container, the measurement may accompany some difficulties. Further, since the production cross section for UCN is so small, the thickness of the He II converter should be relatively large (it amounts sometimes 3 m as exemplified in a device by Golub et al. [19].) in comparison with the SD$_2$ converter. This is another drawback of the He II converter. In fact, a whole size of the UCN source must be designed to be large, which causes difficulty in irradiating the whole converter volume with high flux neutrons.

On the other hand, though the SD$_2$ converter is not an ideal superthermal converter in the sense that UCN loss due to absorption and up-scattering originating from incoherent scattering amplitude of deuteron [20], we can still employ the SD$_2$ as a superthermal converter if we choose a preferable condition, that is, the production rate of UCN can be sufficiently faster than the up-scattering rate and nuclear absorption in the SD$_2$, and the energy distribution of neutrons could be cooled down lower than the SD$_2$ temperature. In this sense, the SD$_2$ converter can also be called a superthermal method. In fact, this method succeeded in producing the world record of UCN density by using a large bulk of SD$_2$ [7].

In 1983 Golub et al. [15] suggested a new type of source for UCN in which the UCN are produced in a thin SD$_2$ film condensed on the walls of a cryogenic container. This new type of source offers some advantages over the bulky SD$_2$ source with possibility of higher performance in view of several practical conveniences such as easy access, small temperature increase due to better thermal conductivity, and so on, which will be described somehow in more detail in Sec. II. In addition, in our proposal, we would actively utilize the high potential energy of SD$_2$ and induces large kick-out velocity of about 5 m/s to UN for realizing high UCN density in a storage vessel.

C. Aim of the present proposal

The aim of the proposal is to primarily verify a new principle of UCN production and storage with a superthermal SD$_2$ which has never been considered so far. Though this method uses a superthermal SD$_2$ similarly to the LANSCE source [7], the method is greatly different each other as far as UCN accumulation is concerned. The LANSCE group uses SD$_2$ in a production vessel connected to a storage vessel whose walls are reflective for UCN. Then, UCN are accumulated in the vessel until an equilibrium UCN density is attained between inside of SD$_2$ converter and outside of it. The characteristic principle is, in more detail, given in Subsec. II B.
On the other hand, our method uses UCN directly preferably emitted with a certain velocity larger than 5 m/s from a surface of SD$_2$ in the direction normal to the surface. Since the lifetime of UCN in the normal SD$_2$ (66% ortho deuterium and 33% para deuterium) at 4 K is estimated to be 4.6 ms [21], the emitted UCN are pulsed if the proton beam is pulsed with a width of, for example, $\sim$5 ms.

When the pulsed UCN emitted predominantly in the normal direction are incident on a wall moving in a reverse direction with respect to the pulsed UCN synchronizing with the proton beam, UCN can penetrate the wall potential and stored in a storage vessel whose walls are coated with a material such as $^{58}$Ni having a sufficiently higher potential than that of SD$_2$. We believe that this method will give us much attractive chance to renew the UCN world record by the LANSCE when we employ almost pure ortho-deuterium as the converter material.

For making this method successful, the choice of a pre-moderator is of importance because a time structure of cold neutrons produced by a pre-moderator should satisfy a time structure for realizing the above method. For this purpose, we propose to check performance of a mesitylene (C$_6$H$_3$(CH$_3$)$_3$) pre-moderator which showed a very efficient performance in producing cold neutrons with a compact size and was resistive against the radiation exposure in our previous work [22–24].

Our proposal is, in other words, summarized as follows: We challenge an attractive task to elucidate characteristics of a compact UCN source by making use of the selected combination of the most efficient pre-moderator geometry and a new concept of a pulsed UCN from the SD$_2$ surface.

II. DETAILED DESCRIPTION FOR THE PROPOSED UCN SOURCE

In this section we describe our proposal. We separated the contents into several subsections to help understanding the subtle points of our proposal. In Subsec. II.A, a general view point is discussed on why the pulsed beams provided from the accelerator facility are advantageous for UCN development relative to CW proton beam or the reactor facility. In Subsec. II.B, we will discuss, in more detail, a basic principle of SD$_2$ superthermal converter and its characteristics for easier understanding our proposal in the adjacent subsection. In Subsec. II.C and II.D, an essence of principle of our proposed UCN source based on pulsed UCN and synchronized moving membrane will be discussed. In the last Subsec. II.E, we discuss characteristics of mesitylene used for a pre-moderator.

A. Advantage of spallation source in pulsed mode

An advantage to use pulsed neutrons produced by spallation reactions with pulsed proton beams from an accelerator has been discussed by many authors [17,26,27]. By pulsing the proton beam and valving off the UCN storage volume from the production volume when the beam is off, one can take advantage of these high neutron densities during the beam-on while by utilizing the period of beam-off to remove heat from the SD$_2$ converter. In this case the maximum UCN density one can produce is limited only by the impulse heating of the converter. In addition to this merit, experiments with the stored UCN can be performed
while the beam is off, thus the S/N will be improved because the backgrounds due to capture gammas accompanying continuously operated reactor sources is expected to be greatly reduced.

**B. Production of UCN by an SD\(_2\) superthermal method**

In this subsection, we give a general description of the production of UCN by a SD\(_2\) superthermal method with emphasis on mechanism of UCN lifetime and maximum available UCN density, which is important in understanding our proposal.

We assume that a SD\(_2\) converter is immersed in a cold neutron. Then, the UCN density produced is expressed by

\[
\rho_u = R \times \tau_{SD},
\]

where \(R\) the UCN production rate of UCN in the SD\(_2\), and \(\tau_{SD}\) is the loss time of UCN in the solid SD\(_2\). In general, the loss time in the normal solid SD\(_2\) is determined by several origins as expressed by

\[
\frac{1}{\tau_{SD}} = \frac{1}{\tau_{phonon}} + \frac{1}{\tau_{D_{abs}}} + \frac{1}{\tau_{H_{abs}}},
\]

where the first term in the right hand side is due to the phonon up-scattering, the second due to the UCN absorption by deuterium, and the last term is that due to the UCN absorption by hydrogen contained in the SD\(_2\) converter as an impurity. Here, the first term in Eq. (2) is most influential term which determines the over-all loss time as discussed in the later part of this section in case of the normal SD\(_2\). It is worthwhile to note that \(\rho\) produced is independent of the size of the converter, though total UCN number produced is proportional to the volume of the converter.

1. **In case of reflective wall**

We treat a specific geometry used by the LANSCE group [7] at first for better understanding our method. We consider, for simplicity, a single closed vessel with walls made of a material with good UCN reflection properties, inside of which a SD\(_2\) with a shape of a film or a bulky volume is located.

Notifying the storage vessel with a volume, \(V\) and the thin layer or bulk of SD\(_2\) with a volume, \(V_{SD} (= \text{area} \times \text{thickness} = A \times d)\) or a same volume of SD\(_2\) is located in the vessel, we estimate the time constant \(\tau\) that the storage vessel comes into equilibrium with the UCN density in SD\(_2\). This time constant is expressed by

\[
\tau = \frac{4V}{v_u A \tilde{\mu}}.
\]

Here, \(v_u\) is the UCN velocity and \(\tilde{\mu}\) is an average loss probability per bounce expressed by

\[
\tilde{\mu} = 4N\sigma_{SD}(v_u)d,
\]
where \( N \) is the number density of the deuterium in the SD\(_2\) and \( \sigma_{SD}(v_u) \) is a loss cross section due to the SD\(_2\) for UCN with the velocity of \( v_u \). Putting Eq. (4) into Eq. (3), the time constant, \( \tau \) is expressed in a simple form

\[
\tau = \left( \frac{V}{V_{SD}} \right) \times \frac{1}{N} \times \frac{1}{v_u \sigma_{SD}(v_u)}.
\]  

(5)

Here, since \( 1/N v_u \sigma_{SD}(v_u) \) in Eq. (5) is equal to the UCN loss time defined by Eq. (2), \( \tau \) is consequently expressed by,

\[
\tau = \left( \frac{V}{V_{SD}} \right) \times \tau_{SD}.
\]  

(6)

2. Numerical values of expected UCN density

In what follows, we can evaluate an analytical form of the \( \rho_u \) deduced assuming that a down-scattering model based on the Debye model is valid and the SD\(_2\) converter is immersed in a cold neutron flux with a Maxwell-Boltzmann distribution at temperature, \( T_n \) by modifying Eq.(10) of Ref. [15]. The result is shown below;

\[
\rho_u = \tau_{SD} \times N \times (2\sigma'_0 \Phi) \frac{m_n}{M} \frac{T_n^2 \theta^3}{\Gamma(T_n, \theta)} \left( E'_{nc} \right)^{3/2},
\]  

(7)

where \( N \) is a number density of the SD\(_2\) \( (\sim 6 \times 10^{22} \text{ atoms/cm}^3) \), \( m_n \) is a neutron mass, \( M \) is a deuteron mass, \( \theta \) is the Debye temperature of the SD\(_2\) \( (\theta = 110 \text{ K}) \), \( E'_{nc} \) is the critical energy for UCN reflection measured in unit of temperature, \( \sigma'_0 \) is the sum of the coherent and incoherent scattering cross section for the SD\(_2\), \( \Phi \) is the total neutron flux, and \( \Gamma(T_n, \theta) \) is given by

\[
\Gamma(T_n, \theta) = 2(\theta)^{7/2} \sum_{n=0}^{\infty} \frac{(-\beta \theta)^n}{n!(2n + 7)},
\]  

(8)

with

\[
\beta = \frac{3 m_n}{2 M \theta} + \frac{1}{T_n}.
\]  

(9)

We take \( E'_{nc} = 3 \text{ mK} \) (Beryllium cut-off) and for SD\(_2\),

\[
\sigma_{SD} \sim 0.22 \text{ barns},
\]  

(10)

and

\[
\sigma'_0 = 7.6 \text{ barns},
\]  

(11)

then we find by using Eq.(3)-(6),

\[
\rho_u \sim 125 \times 10^{-11} \Phi_0 \text{ [UCN/cm}^3\text{]} \text{ (at } T_n = 25 \text{ K)}. 
\]  

(12)
According to our Monte Carlo calculation, cold neutron flux $\Phi_0$ is $\sim 10^{10}$ for the mesitylene pre-moderator in case of the proton beam of $1 \mu A$, which results in

$$\rho_u \sim 12.5 \text{ [UCN/cm}^3\text{]}.$$  \hspace{1cm} (13)

The above calculation was performed with the normal SD$_2$ assuming that the UCN loss due to the up-scattering is ignored. If the effect of the UCN up-scattering is taken into account, $\rho_u$ becomes small by an order of magnitude as discussed in the next Subsec. II B 4

3. $T_n$ dependence of UCN density

Yu et al. [16] treated the UCN density more realistically by using realistic phonon spectra. The realistic models provided important information on the temperature dependence of the down-scattering cross section in designing a pre-moderator. The results of the calculation of $\rho_u$ plotted as a function of $T_n$ are shown in Fig. 1. The noteworthy results are that the density reaches a maximum value at $\sim T_n \sim 30 \text{ K}$. 
FIG. 1. $\rho_u$ [UCN/cm$^3$] for SD$_2$ converter plotted as a function of neutron temperature, $T_n$ for various models in unit of $\Phi_0 \times 10^{-11}$

4. Temperature dependence of $\tau_{SD}$

It was experimentally found that the loss time, $\tau_{SD}$ of UCN in the SD$_2$ is mainly determined by the up-scattering due to the para deuterium composition at low temperature ($\sim$ 4 K) [7]. This behavior was theoretically ensured by Liu et al. as shown in Fig. 2, where the temperature dependences of the cross sections for not only the para but also the ortho deuterium were plotted [21]. Fig. 2 also shows that the para deuterium has a temperature independent spin-relaxation channel (see the dash-dagger curve denoted $J = 1 \rightarrow 0$). The conversion energy of 7 meV can be released to UCN, resulting in a loss time of 4.6 ms in a normal deuterium, in which 33% para deuterium is contained. This up-scattering term was not taken into account in the derivation of Eq. (12). In other words, Eq. (12) corresponds to an ideal value for pure ortho SD$_2$ and the loss time is $\tau_{SD} \sim 300$ ms. On the other hand, since $\tau_{SD}$ for the normal ortho SD$_2$ at 4 K is 4.6 ms [21], an attainable UCN density reduces orders of magnitude as shown by

$$\rho_u = 0.19 \text{ [UCN/cm}^3\text{]}, \quad (14)$$

where the SD$_2$ temperature is 4 K, and the proton beam intensity is 1 $\mu$A. However, if we choose to use the ortho SD$_2$ converter with a minor content of the para deuterium of 2.5%, $\tau_{SD}$ is increased up to 30 ms. Then, an attainable UCN density is increased to

$$\rho_u = 1.25 \text{ [UCN/cm}^3\text{]}, \quad (15)$$

where the SD$_2$ temperature is 4 K, and the proton beam intensity is 1 $\mu$A.
FIG. 2. UCN up-scattering cross section vs. temperature of SD$_2$. The one phonon annihilation cross section in an ortho D$_2$ solid (solid curve) and in a para D$_2$ solid (dashed curve) are plotted. The dashed dagger line is the temperature independent UCN up-scattering cross section involving J=1→0 relaxation not coupled to phonons in a para D$_2$ solid.

C. Pulsed UCN

According to the discussion of the previous Subsec. II B, the UCN produced in normal SD$_2$ have a time constant, $\tau_{SD} \sim 4.6$ ms assuming that the lifetime is predominantly determined by the para deuterium. When UCN reach on the SD$_2$ surface, they will be emitted with velocities larger than 5 m/s because of the potential of SD$_2$ felt by UCN. Though the angular distribution of UCN normal to the SD$_2$ surface are not well known, it may not be a bad approximation that UCN are preferably emitted to the normal direction. Further, UCN are in a pulsed scheme emitted from the SD$_2$ surface in time if we use a pulsed beam. This is an excellent characteristics which is never seen in other converters such as He II. The directivity and pulse of UCN are very convenient for accumulating in a storage vessel by using a moving membrane discussed in Subsec. II D.

D. Moving membrane

A principle of this method is intuitively described below. We assume that UCN with a velocity $v$ in a production vessel hits a membrane whose critical velocity is $v_c$ ($v_c \geq v$) as
shown in Fig. 3.

\[ a) \text{Rest membrane} \quad b) \text{Moving membrane} \]

![Diagram showing a schematic picture of a moving membrane](image)

**FIG. 3.** Schematic picture showing a principle of a moving membrane.

1. **Principle of moving membrane**

If the membrane is in rest (a) in Fig. 3), the UCN cannot penetrate the membrane because \( v \) is less than the critical velocity of the membrane. On the other hand, if the membrane moves with a velocity \( u \) (b) in Fig. 3), a part of UCN component which has a velocity satisfying the following un-equality can penetrate the membrane and get into a storage vessel,

\[ v_\perp + u \geq v_c, \]

where \( v_\perp \) is a velocity component to the \( z \) direction. In fact, from SD\(_2\) surface in parallel to the flat membrane, UCN will be emitted with normal velocity component, \( v_\perp \) accelerated by about 5 m/s over that inside the SD\(_2\). Suppose that we use \(^{58}\text{Ni} \) (\( v_c = 8.3 \) m/s) as a surface of a moving membrane and a storage wall. If we operate the membrane with a velocity 3.3 m/s, UCN with a velocity range from 5 to 8.3 m/s can be stored in a storage vessel. This is our basic idea of a moving membrane.

To realize this principle we design a moving membrane operated by a driving mechanism with a time sequence as shown in Fig. 4.
FIG. 5. Velocity gain of UCN in the vessel due to the collision with moving wall.

Another important aspect of this model is that UCN stored in the storage vessel are expected to gain a kinetic energy (or velocity) as a result of total reflection by the membrane moving with a velocity \( u \). The velocity gain of UCN is simply estimated as follows. In our present one-dimensional model, a velocity gain of UCN with a velocity, \( v \) bounced off the moving membrane per one bounce is 2\( u \). The number of UCN bouncing off the membrane during a short time, \( \Delta t \) is given by \( \Delta t \times v/2L \). Therefore, velocity increase of UCN during the time, \( \Delta t \) is given by

\[
2u \times \frac{\Delta t}{2L} = v \times \frac{\Delta tu}{L} = v \times \frac{\Delta L}{L}.
\]  

This result indicates that velocity gain due to collision of UCN with the moving membrane is independent of the velocity \( u \), i.e., the velocity of the membrane and is determined only by the volume change of the storage vessel. From this result, it is concluded that a velocity gain can be kept small by choosing \( \Delta L/L \) as small as possible. Of course, we must notice that in a such small ratio for \( \Delta L/L \), a large number of repetitions, or a long operation time in total, should be required until attaining a saturated UCN density in the storage vessel.

In the following, we will see the appropriateness of the above results from the principle of the thermodynamics.
3. Thermodynamical consideration

Here, we present a short consideration on stored UCN dynamics by considering it as analogy with the rarefied gas dynamics in a closed vessel.

An (kinetic) energy increase of UCN, $\Delta E$ is estimated from Eq. (17), and is given by

$$\Delta E = \frac{m}{2} v^2 \left( 1 + \frac{\Delta L}{L} \right)^2 - \frac{m}{2} v^2$$

$$\sim 2 \times \frac{m}{2} v^2 \frac{\Delta L}{L}$$

$$= 2 \times E \frac{\Delta L}{L}, \quad (18)$$

where $E$ is a total kinetic energy of UCN gas. Here, $\Delta L/L$ is assumed to be infinitesimal and their higher order terms can be neglected. The origin of the factor, 2 in the right hand side of Eq. (19) is explained by the thermodynamical consideration as shown below.

An ideal gas obeys the Boyle-Charles's equation,

$$PV = kE,$$  \hspace{1cm} (20)

where $E$ is a total gas kinetic energy and $k$ is a constant depending on the dimension, $k=2/3$ for the three dimensions, and $k=2$ for the one dimension. Therefore, an energy change, $\Delta E$ is given by

$$\Delta E = \frac{1}{k} \times \Delta (PV)$$

$$= \frac{1}{k} (\Delta P \cdot V + P \cdot \Delta V)$$

$$= \frac{1}{k} PV \left( \frac{\Delta P}{P} + \frac{\Delta V}{V} \right)$$

$$= E \left( \frac{\Delta P}{P} + \frac{\Delta V}{V} \right)$$

$$\sim 2 \times E \frac{\Delta L}{L}. \quad (21)$$

As demonstrated in the derivation process of Eq. (21), the factor, 2 is a natural consequence of the thermodynamics that the work exerted by the membrane for the UCN gas is not restricted only to compression, but also to pressurization of the UCN gas.

4. Optimization of time sequence

As described previously, Fig. 4 shows one cycle of a time sequence needed for our synchronized storage method. We try to give numerical values for the times denoted by $T_1, \ldots, T_4$ for the proposed experiment. $T_1$ is a beam-on time. Since the UCN loss time in the SD$_2$ converter is 4.6 ms, it is suitable to choose $T_1 \sim 5$ ms. $T_2$ is a time of flight for UCN produced in the SD$_2$ converter to reach the membrane surface. If we choose the
distance between the SD$_2$ converter and the membrane to be 5 cm, T.O.F is 10 ms for UCN with 5 m/s and 5 ms for UCN with 10 m/s. Taking the beam-on time (~5 ms) into account, T$_2$ should probably exist in 0~5 ms. T$_3$ is a moving time of the membrane. Since the time length of the pulsed UCN is ~15 ms, T$_3$ should be at least 15 ms. In this case, displacement is

$$330 \times 15 \times 10^{-3} = 4.95 \text{ cm.}$$

(22)

T$_4$ is a time needed for the return process and should be chosen so that T$_3 \ll T_4$. Then it is not so bad to choose T$_4 \sim 80$ ms. In conclusion, a cycle time is T$_1$+T$_2$+T$_3$+T$_4$ = 5+0+15+80 = 100 ms. This time sequence gives a beam duty factor of 5%.

5. Time lengths needed for accumulation, storage, and detection of UCN

Here, we estimate the times needed for accumulation, storage, and detection of UCN. Their time sequence is schematically shown in Fig. 6.

![FIG. 6. Time sequence for accumulation, storage and detection of UCN.](image)

A specific time for accumulation and efflux of UCN in the storage vessel is called the efflux time and defined by

$$T = \frac{4V}{Av},$$

(23)

where V is a volume of storage vessel, A is an area of hole of the vessel through which UCN are introduced from the outside or efflux to the outside of the vessel. At first, we evaluate
the accumulation time, \( T_A \). If we choose \( A = 100 \text{ cm}^2 \), \( v = 500 \text{ cm/s} \), and \( V = 1000 \text{ cm}^3 \), then Eq. (23) results in \( T = 80 \text{ ms} \). Assuming the equilibration time is \( \sim 20T = 1.6 \text{ s} \), it is reasonable to choose \( T_A = 5 \text{ s} \).

The UCN lifetime in the storage vessel is determined by the wall loss and the natural lifetime of UCN, and usually longer than 100 s. To check the quality of the SD\(_2\) converter, it is favorable that the measurement is performed with different values of \( T_S \) around 40 s, for example.

Finally, we estimate the detection time of UCN. To detect UCN, we open shutter with a small hole with an area, \( A \) in the storage vessel. In this case, the efflux time is given also by Eq. (23). Assuming that \( A = 10 \text{ cm}^2 \), we can obtain \( T = 800 \text{ ms} \). By taking the time needed for UCN transportation into account, we set \( T_D = 5 \text{ s} \).

### E. Pre-moderator

A special attention should be paid for a high density UCN source that uses a converter method in conjunction with a spallation neutron source. Since the primary advantage of the spallation source is its compactness, a selection of suitable pre-moderator to meet this condition is of particular importance. From this point of view, hydrogen compounds such as methane and polyethylene have been offered as an efficient pre-moderator. Further, from the viewpoint of the time structure in the neutron slowing-down, cold neutron buildup and their finally decay out, most of hydrogenous moderators show the slowing-down and cold neutron buildup time of a few tens \( \mu \text{sec} \), and the decay time of 150-200 \( \mu \text{sec} \). These time constants are sufficiently short comparing to our supposed time sequence and therefore we can expect efficient performance and speedy time response of the hydrogenous pre-moderators for our concept by the simple time control of the proton beam.

Among these hydrogen compounds, solid methane has shown the highest cold neutron performance. However, since the severe decomposition of methane due to the radiation exposure cannot be avoided, the solid methane is not the best solution as the pre-moderator for a long time use under severe radiations.

The mesitylene pre-moderator was developed so as to fit a compact sized pulsed neutron facility [22–24]. The mesitylene is a kind of methyl-benzene, and characterized by a much effective moderation performance for cold neutrons owing to the methyl groups, the high radiation resistance of the benzene ring, and an easy handling with a less hazard.

According to the performance of the mesitylene as a pre-moderator investigated at various temperatures from room temperature to 25 K [22,24], the cold neutron flux at the neutron energy of 1−5 meV seems to show an increase even at temperatures lower than 25 K, as shown in Fig. 1 for a typical case of the neutron energy \( E = 2.5 \text{ meV} \), i.e. the most important region for the solid deuterium converter. This striking result can never be understood in terms of the presence of the lowest \( \text{CH}_3 \) rotational and vibrational peak at \( E_x = 6.9 \text{ meV} \) [25]. In other words, this result strongly implies a presence of the unknown vibration or any other mechanism with the energy lower than this peak.

In addition, from a practical point of view, the above experimental result strongly suggests a further increase of the cold neutron flux might be expected if the temperature is
lowered than 25 K. In other words, a substantial improvement of the performance as pre-moderator may be possible in a temperature region lower than 25 K.

FIG. 7. Temperature dependence of neutron flux measured at $E = 2.5$ meV in mesitylene moderator
If this is experimentally ensured, this will provide a striking impact on the improvement of the UCN sources based on the spallation method now working or being designed in the world. In fact, since the world record of the highest UCN density attained by the UCN source at LANSCE [7] is based on the polyethylene pre-moderator which has less pronounced performance than the mesitylene working at $T = 25K$, a remarkable improvement of the UCN density could be expected if the polyethylene is replaced by the mesitylene working at $T \leq 25K$. The neutron flux in the energy region $E \leq 3$ meV being approximately linear to the energy $E$, Fig. 7 also suggests an increase of the flux at $E = 1$ meV at lower temperatures than 25K. The neutrons with the energy 1 meV are used also as incident neutrons for the superthermal helium converter. From this result, we must measure the temperature dependence down to 10K on the TOF spectra and intensities for cold neutrons from a mesitylene pre-moderator with the thickness of 4.5 cm which corresponds to an optimized thickness by our Monte Carlo simulation as shown in Fig. 8.

![Neutron Flux at 5meV](image)

**FIG. 8.** Result of Monte Carlo simulation for the cold neutron yield at the spectral peak $E\sim5$meV from the mesitylene pre-moderator ($T = 22K$).

Needless to say, an unexpected cooling mechanism for the mesitylene at low temperature will open a new physics field. Therefore, our study on the mesitylene pre-moderator will be instructive not only for the practical application but also on the fundamental material physics.
III. EXPERIMENTAL EQUIPMENT

We have designed a UCN source to examine the validity of the proposed principle, i.e., a synchronized SD$_2$ storage method with mesitylene pre-moderator. For this purpose, we have revised a He cryostat which was used at KUR in the past. A schematic view of the UCN production system designed is shown in Fig. 9.

![Schematic view of UCN production system](image)

**FIG. 9.** Schematic view of UCN production system

We design a closed pre-moderator chamber, a closed solid deuterium converter section, and a UCN storage vessel separated by a moving membrane, so that these three sections could be installed in a single cryostat. This equipment is also designed so that the TOF measurement for the performance measurement of the pre-moderator. A special care was paid to designing a deuterium gas handling system. The gas handling system allows a fine regulation of gas flow, and monitoring the flow rates and pressure to estimate the SD$_2$ thickness. For detecting UCN we use a thin-walled $^3$He counter specially prepared for UCN experiment and used at KUR.
IV. EXPERIMENT

In this chapter, we present a status of preparation, setup, the expected yields and spectra, and beam requirement.

A. Status of preparation for experiment

A target system consisting of a vacuum chamber and a movable Pb target with a 5cm in diameter and 20cm in length is already ready for use. The performance of a liquid He cryostat for deuterium converter condensation, solidification and cooling nearly down to 4 K, and also for cooling mesitylene pre-moderator down to 10K is now tested. A set of BF$_3$ neutron detector and data taking system for cold neutron TOF experiments on the pre-moderator are in preparation. Containers of mesitylene and a flight tube with about 6 m in length are designed to be machine-out. The new proton beam line in the East Experimental Hall in RCNP has just been completed and passed the operation test in the early October, 2001.

Therefore, we consider that our proposal is almost ready to start.

B. Expected yields and spectra

1. Experiment of UCN storage

According to our previous experience, the maximum beam intensity should be less than 3 nA with the target and cryogenic system that we are going to use in this proposal from the radiation protection reason.

On the other hand, in Sec. II we have discussed the detail of the experimental method to store UCN in the storage vessel. We found from the discussion on the optimized time sequence that the beam-on time is 5 ms and a cycle time is 100 ms with a duty factor of 5%.

This indicates that we can use a pulsed proton beam with a beam current of 60 nA.

In Subsec. II B 4, we estimated UCN density to be 0.19 UCN/cm$^3$ by using Eq. (14) for the normal SD$_2$ with the proton beam of 1 $\mu$A. For 600 nA for the synchronized UCN filling followed by our proposed measuring scheme with $T_M = T_S + T_D = 45$ s after filling for $T_A = 5$ s, the UCN density becomes about 0.12 UCN cm$^{-3}$. We assume the volume of the store vessel is one liter. Then, the total number of UCN in one liter vessel is about 120 UCN/filling, and a beam running for about 24 hours, or about 1,700 cycles with 5 s filling and 45 s storage-and-counting by using a UCN counter with the detection efficiency assumed to be 0.5 will enable us to get a counting number of about 10$^5$, which should give us enough statistics for distinguishing UCN counts from the background counts with our specially prepared UCN detector having the characteristics of very low background counting rate far below 0.01 cps including cosmic ray contributions, which is two orders of magnitude smaller than the true event.

Among the 4 days requested, one day will be consumed for tuning of the proton beam and the experimental device including the UCN detector, and other 3 days will be used for optimization of the time sequence, $T_n$ dependence, and the SD$_2$ temperature dependence.
2. Experiment of mesitylene pre-moderator

In the following, we estimate the beam time: The yield (counts/sec.) of cold neutrons to be detected at the end of the flight tube (~6m) for the incidence of a 3 nA proton beam is estimated. In the TOF measurement, 0.048 neutrons/s are counted for the repetition period of 40 ms, where it is assumed that the duty factor of the beam is 0.5%, and the effective area of a neutron detector is 20 cm². Assuming a necessary statistics is 1000 counts/flight-time channel with a flight-time step of 0.1 ms, the accumulation time is about 2 hours. If one observes the TOF spectra at 5 different temperatures, 10 hours are necessary for the whole measurements. Assuming that another 1 hour is necessary between each run for changing the temperature after each run, 4 hours should be added. Consequently, 14 hours are needed. Another 10 hours are necessary for beam transportation. The same experiment should be done by changing the thickness of the mesitylene pre-moderator. The total running time is, then, 2 days.

V. FUTURE OUTLOOK

One of the important subjects we did not touch in this proposal is an up-scattering of UCN by a para deuterium. Since the up-scattering by the para deuterium seriously contributes to greatly reducing the expected UCN density [21,7], we must diminish the content of the para fraction in $\text{SD}_2$ converter. For this purpose, we will design a practical UCN source so that the para fraction may be controlled. This will be done by converting the deuterium to a near thermal equilibrium ortho/para ratio in an iron hydroxide filled cell cooled to a temperature at or slightly below the triple point; an equilibrium value of content of para deuterium (33%) would reduce to 2-3 %.

After constructing a practical UCN source and examining the performance, we will start the search of fundamental physics with the UCN intensity for the source as soon as possible.
REFERENCES

[12] Y. Masuda et al., approved as the RCNP project.