Journal of Neutron Research 24 (2022) 167–177
DOI 10.3233/JNR-220008
IOS Press

The source for ultra-cold neutrons at the FRM II

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Abstract. At the Forschungs-Neutronenquelle Heinz Maier-Leibnitz (FRM II) of the Technical University of Munich (TUM) a new source for ultra-cold neutrons (UCN) with a solid deuterium converter is currently under construction. This summary paper shall give an overview of the project and its current status. Research results concerning converter preparation, para-to-ortho conversion, radiation effects and neutron transport, which have been achieved in the last years, are presented and their relevance and transferability for the design of a future UCN source at the European Spallation Source (ESS) are discussed.

Keywords: Ultra-cold neutrons, solid deuterium converter, para-to-ortho deuterium conversion, ultra-cold neutron transport, ultra-cold neutron source at ESS

1. Introduction

Precision experiments with ultra-cold neutrons [37,63], such as the search for a possible electric dipole moment (EDM) of the neutron [1,46,54] or the measurement of the lifetime τ_n of the free neutron [6,16,25,45,47,52,56], require high UCN densities. Stronger UCN sources are presently developed worldwide, based on the principle of superthermal UCN production, using cryo-converters made of solid deuterium (sD_2) or superfluid helium [4,10,18,23,24,30–32,34,35,38,39,48,57,58,72,73]. At the FRM II a UCN source with a sD_2 converter and sH_2 premoderator, placed in a distance of ∼60 cm from the central fuel element inside the horizontal, through-going beam tube SR6, is currently under construction. It is supposed to generate UCN densities of 10^3–10^4 cm^{-3} in up to four connected experiments.

2. Design of the UCN source at the FRM II

At the FRM II the through-going beam tube SR6 is currently unused. It has one beam port exit at the east side of the experiment hall (called A-side), and one beam port exit in the neutron guide tunnel at the west side (called B-side). This offers the possibility to install all necessary cryogenic supply lines from the B-side, and thus the A-side is completely free for undisturbed extraction of the UCN to connected experiments. The position and the layout of the UCN source is depicted in Fig. 1.

The central part of the UCN source is the converter vessel, a double-walled toroidal-shaped aluminium cap piece, which is cooled by a continuous flux of a closed supercritical helium cooling loop. The necessary cooling power of up to 1.0 kW at 5 K is supplied by two cold boxes (AirLiquide Helial 2000) to the closed supercritical He-loop. The converter contains 12.5 mol of solid hydrogen (sH_2) as premoderator (volume ∼ 250 cm^3) to precool the incoming thermal neutron flux ($1.1 \cdot 10^{14}$ $cm^{-2}s^{-1}$) to an effective neutron temperature of ∼40 K. The sD_2 UCN converter (maximum amount 12.5 mol) is frozen to the outer surface of the converter vessel by resublimation

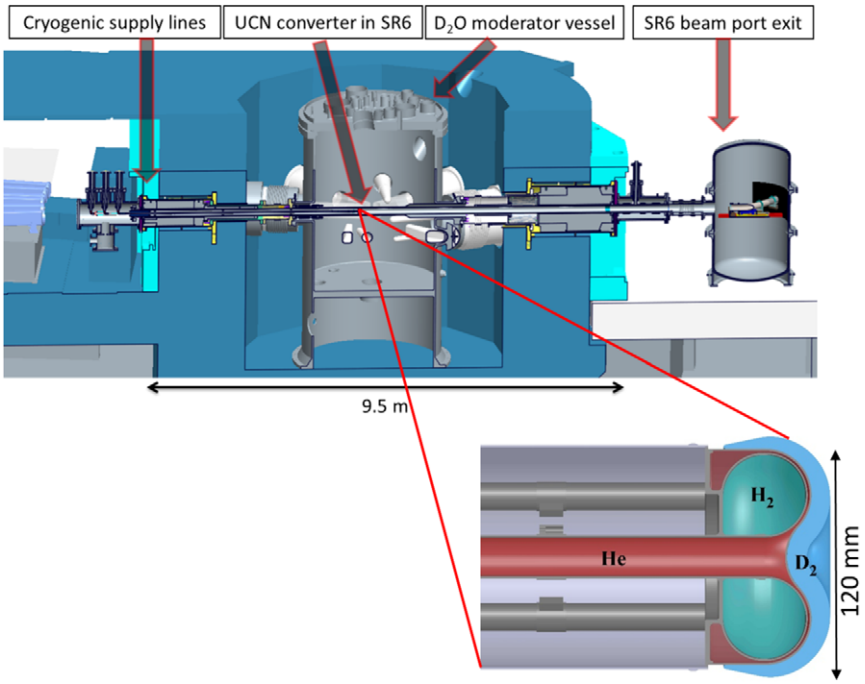

Fig. 1. **Top:** vertical cut view of the heavy water vessel and the surrounding concrete shielding (indicated by blue color) of the FRM II along the SR6 beam axis. The cryogenic supply lines for the UCN converter are installed from the left (called B) side. UCN can be extracted to the opposite SR6 A-side through a safety vessel to connected experiments. The UCN converter is placed approximately in the middle of the SR6 beam tube, in a distance of ∼60 cm from the central fuel element of the FRM II. **Bottom:** vertical cut view of the UCN converter, a double-walled, toroidal-shaped aluminium vessel. Both walls are cooled by a closed supercritical helium cooling loop (indicated by red color) to a temperature of ∼5 K. Inside the vessel solid hydrogen as premoderator is frozen out (indicated by green color), and the solid deuterium converter (indicated by blue color) is resublimated from gas to solid state to the outer wall of the aluminium vessel.

of D_2 gas to the solid phase. The premoderated incoming neutrons can enter the sD_2 converter, where they induce solid-state excitations (mainly phonons) of the crystal lattice or rotations of the molecules. Solid ortho-deuterium has exited states in the energy range of 2–20 meV. If the kinetic energy of an incoming neutron equals the energy of a solid-state excitation, the incoming neutron can lose practically its total initial energy, and is converted into the energy regime of ultra-cold neutrons [19]. The UCN generated by this process can leave the sD_2 converter, and are guided to the SR6 beam port exit in the experiment hall, and fed into connected experiments. The expected UCN flux density at the SR6 A-side beam port exit is $6 \cdot 10^5$ cm^{-2}s^{-1} within the energy range of 100 neV–230 neV and a beam aperture of 100 cm^2 [17].

3. Solid deuterium as UCN converter material

3.1. Para-to-ortho conversion

Since the inelastic up-scattering cross-section for UCN on para-deuterium $\sigma_{\text{para}} \cdot v \approx 250$ barn · m/s is about a factor of 150 higher than that of ortho-deuterium $\sigma_{\text{ortho}} \cdot v \approx 1.6$ barn · m/s at 5 K [36], an efficient para-to-ortho conversion is a prerequisite to keep the UCN lifetime in the converter material close to its optimum value at about 70 ms. Natural thermal conversion of para-to-ortho state in sD_2 takes months [60]. Therefore, we use OXISORB® [67] (chromium oxide bound in silica grains) as paramagnetic catalyst to accelerate the spin-flip process in a separate cryogenic cell. A detailed description of the process can be found in ref. [12]. The OXISORB® is cooled

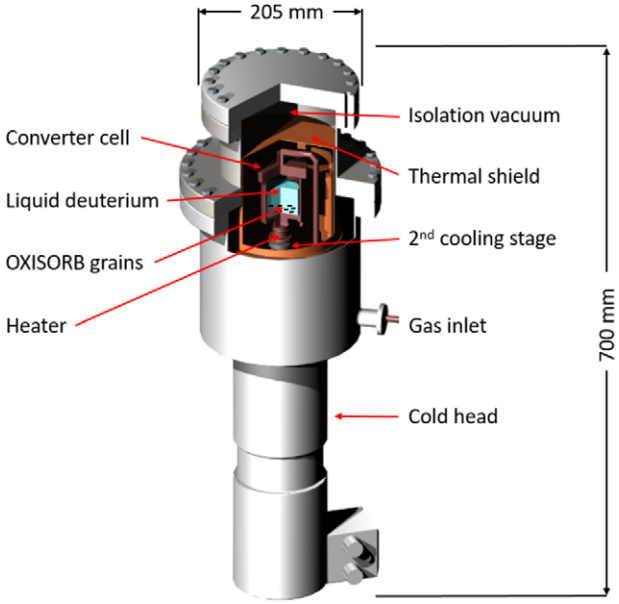

Fig. 2. Scheme of the para-to-ortho converter using the surface contact of liquid deuterium with OXISORB® grains. The converter cell is mounted on the 2nd cooling stage of the cryo-cooler, while the thermal shield is cooled by the 1st stage.

by a separate two-stage Gifford–McMahon cryo-cooler, with a cooling power of 0.5 W at 4.2 K. It turned out that conversion temperatures just above the triple point ($T_{con} = 20$ K) are optimal for our purpose, since deuterium from the liquid phase can be boiled to provide close contact of the D_2 molecules with the surface of the catalyst. With this method it could be shown that the conversion rate can be enhanced up to values of 0.35 h^{-1} [17]. The residual population of the para state is characterized by the para-to-ortho transition energy ($E_{10} = 7.5$ meV) and the Boltzmann factor $\exp(-E_{10}/(k_B \cdot T_{con}))$, resulting in a para-$D_2$ concentration of about 1% in the low-temperature limit of sD_2. The ratio of para-D_2 to ortho-D_2 is determined off-line by means of a Raman spectrometer, using a small sample of deuterium gas which is extracted by warming up the frozen deuterium. A para-concentration of $< 2\%$ could be confirmed in all measurements. A scheme of the converter unit is shown in Fig. 2.

3.2. Converter preparation

The preparation of an optically transparent sD_2 converter crystal is an important task to avoid additional losses of UCN due to scattering on inhomogeneities, such as grain boundaries. Solid deuterium can be frozen out either from the liquid or directly from the gaseous phase via resublimation. Due to the horizontal geometry of the FRM II UCN source, freeze-out via resublimation is the only possible way for preparation of the sD_2 converter. To study all important parameters which influence the crystal quality, such as temperature and temperature gradients, pressure, speed of freeze-out and incoming mass flow of gaseous deuterium, an aluminium sample cell with an inner volume of $30 \times 30 \times 30$ mm^3 equipped with windows made of sapphire glass was mounted on a two-stage Gifford–McMahon cryo-cooler. With a heater at the second stage and a LakeShore temperature controller it was possible to adjust the cell temperature from 8 K to 300 K. It was possible to create optically transparent crystals by resublimation at temperatures < 10 K when the incoming flow of deuterium gas was $\dot{V} \leqslant 30$ sccm. Pictures of sD_2 crystals generated under different conditions are depicted in Fig. 3. A detailed description of the setup and the freeze-out procedures can be found in ref. [41].

After these preliminary investigations, a $1 : 1$ prototype of the converter vessel of the FRM II UCN source has been cooled with liquid helium down to a temperature of 5 K in a test setup and the freeze-out process via resublimation has been studied. Also here optically transparent deuterium crystals could be produced when the incoming gas flow was $\dot{V} \leqslant 30$ sccm. A detailed description of these experiments can be found in ref. [68].

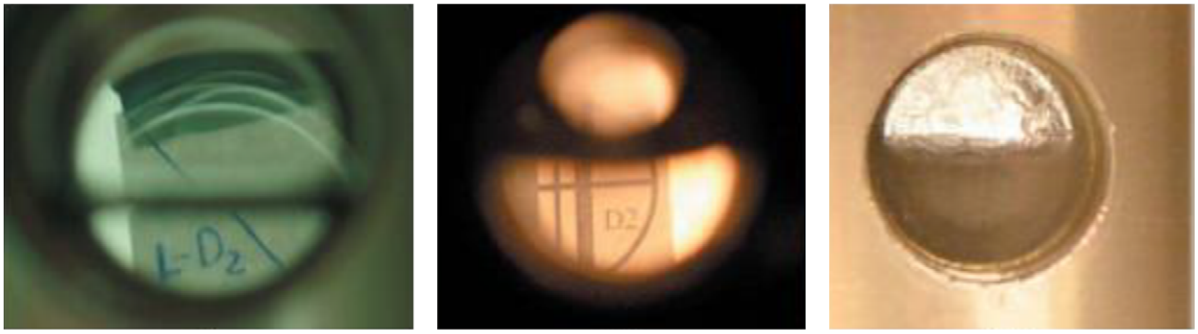

Fig. 3. **a)** Solid deuterium frozen out from the liquid phase. **b)** Solid deuterium frozen out via resublimation from the gaseous phase at $T = 8$ K with an incoming gas flow of $\dot{V} \leqslant 30$ sccm. **c)** Solid deuterium frozen out via resublimation from the gaseous phase at $T = 11$ K with an incoming gas flow of $\dot{V} \gg 30$ sccm.

3.3. Radiation-assisted para-to-ortho conversion

Besides by catalytic materials, the ortho-fraction in deuterium can also be increased by irradiation. It is well established that atomic deuterium produced by radiation effects is responsible for the $J = 1 \rightarrow 0$ transition in deuterium [7,13,50,59,62]. Atomic deuterium is paramagnetic, because of its unpaired single electron. Thus, it provides the necessary magnetic moment to flip nuclear spins of the surrounding molecules.

Any kind of radiation, be it neutron, γ or β radiation, will ionize the deuterium either directly through electromagnetic interactions or indirectly by first colliding with an atom, which then causes ionization. The main ionization processes in deuterium are [62]

$$\mathrm{D_2} + 15.43\ \mathrm{eV} \rightarrow \mathrm{D_2^+} + e^- \tag{1}$$

$$\mathrm{D_2} + 18.08\ \mathrm{eV} \rightarrow \mathrm{D} + \mathrm{D^+} + e^- \tag{2}$$

$$\mathrm{D_2} + 31.67\ \mathrm{eV} \rightarrow 2\mathrm{D^+} + 2e^-. \tag{3}$$

Lower energetic particles may also excite the electronic system of the deuterium without ionizing it, leading to the reactions

$$\mathrm{D_2^*} \rightarrow \mathrm{D_2} + h\nu \tag{4}$$

$$\mathrm{D_2^*} \rightarrow 2\mathrm{D} \tag{5}$$

$$\mathrm{D_2^*} \rightarrow \mathrm{D^*} + \mathrm{D} \tag{6}$$

$$\mathrm{D_2^*} \rightarrow 2\mathrm{D^*}. \tag{7}$$

The reactions (1)–(7) are leading to 1.14 directly formed atoms per created ion pair. Since $\mathrm{D_2^+}$ is only stable in high vacuum, it will convert to $\mathrm{D_3^+}$ via the reaction

$$\mathrm{D_2^+} + \mathrm{D_2} \rightarrow \mathrm{D_3^+} + \mathrm{D} + 2.25\ \mathrm{eV}$$

This leads to another free deuterium atom, now formed indirectly. The $\mathrm{D_3^+}$ ions will at some point recombine with an electron according to the reaction

$$\mathrm{D_3^+} + e^- \rightarrow \mathrm{D_2^*} + \mathrm{D} \rightarrow 3\mathrm{D} \rightarrow \mathrm{D_2} + \mathrm{D}$$

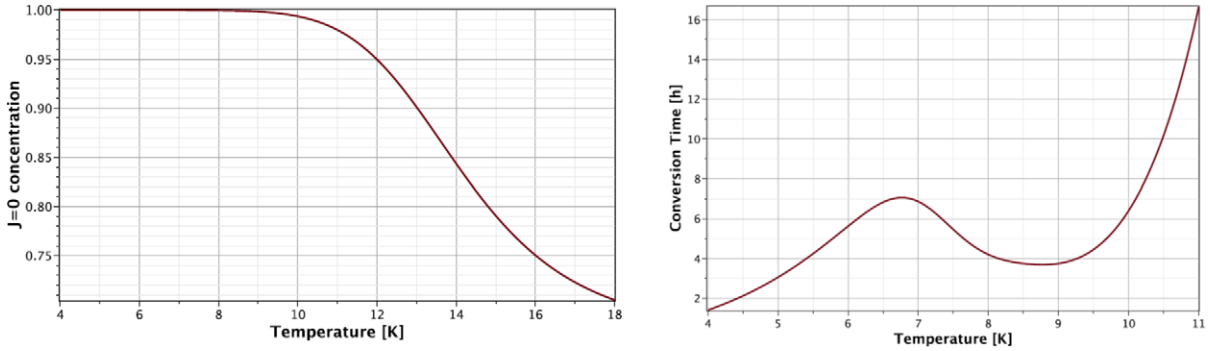

Fig. 4. **Left:** the $J = 0$ concentration for the $t \to \infty$ case in the FRM II UCN source depending on the temperature. Up to 9 K the deuterium will be completely in its ortho state. At higher temperatures the ortho concentrations drop rapidly towards the high temperature equilibrium due to the recombination of deuterium atoms. **Right:** calculated conversion time constant for a deuterium crystal under irradiation with a dissociation rate of $2.84 \cdot 10^{-6}$ s^{-1} as expected for the FRM II UCN source. Both figures from [69].

This reaction is the main source of atomic deuterium under irradiation. On average there will be 5.1 ± 0.3 atoms created per ion pair. Even though this value has been determined in hydrogen gases, experiments have shown that it still holds true in solids. Taking into account the molecular break-up of deuterium, transitions between para and ortho deuterium, and recombination of deuterium atoms to molecules, a differential equation can be set up to calculate the $J = 0$ content in deuterium under irradiation. Its solution, for the conditions of the FRM II UCN source, has been proven experimentally, see ref. [69]. As a result, Fig. 4 shows the ortho concentration in the FRM II UCN source for the $t \to \infty$ case as well as the expected conversion time constant. One can see that up to 9 K the deuterium will be almost completely in its $J = 0$ state. For higher temperatures there is a sharp drop almost down to the high-temperature equilibrium values of the ortho concentration. The atoms recombine too quickly and the newly formed molecules will have the high temperature equilibrium ortho concentration. At temperatures below 9 K the $J = 1 \to 0$ conversion rate is higher than the recombination rate of the atoms, while at higher temperatures the recombination rate exceeds the $J = 1 \to 0$ rate by several orders of magnitude, e.g., by five orders at 12 K.

4. UCN transport

The efficient transport of UCN from their source to the experimental site is a major issue for various kinds of precision experiments. Neutron guides often have to transport the UCN several tens of meters with acceptable losses. At the FRM II a site dedicated to investigations with UCN is foreseen in an external experimental hall about 40 m away from the new UCN source. The site offers an environment adequate for precision experiments, in terms of disturbing mechanical vibrations, magnetic fields, electromagnetic noise and radiation.

UCN can be guided by total reflection in tubes either made of a suitable neutron reflecting material or coated with such a material. The transport is accompanied by many wall reflections and hence the loss probability per wall collision is an important quantity [3,11,14,15,28,33,40,42,43,49,53,61,64,66,71], as well as transport losses by diffuse reflection. As known, for a rough surface the probability of diffuse scattering is approximately given by $\sim(\Delta d/\lambda)^2$ [42,49,64], where Δd is the average surface roughness and λ the UCN wavelength. Therefore the roughness should be in the order of 1 nm or better, to decrease the probability of diffuse reflection to below 1%. In addition, for a multi-purpose UCN guide system the neutron optical potential V_F should be as high as possible, especially for a solid deuterium UCN source ($V_F(sD_2) \approx 100$ neV) as it is planned at the FRM II.

One possibility to produce UCN guides with low surface roughness and high neutron optical potential is to fabricate foils via the replication technique (replica guide), which is already used for many years at the Institut Laue-Langevin (ILL) UCN facility [65]. The surface exposed to the UCN is a copy of a float glass surface, onto

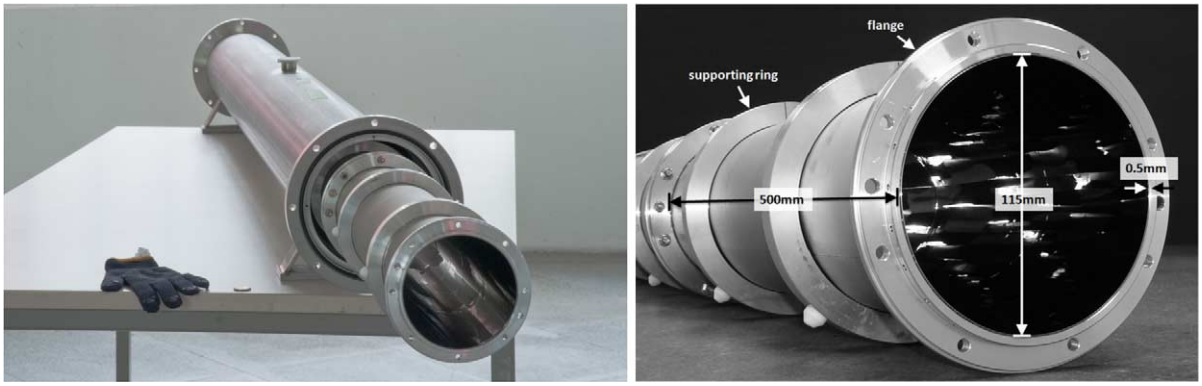

Fig. 5. **Left:** a part of a replica foil with its supporting aluminium ring and its flange at the end of the foil is shown. **Right:** photograph of two connected replica foils with their dimensions.

which a Ni-alloy is deposited by sputtering, reinforced by natural nickel by a galvanic process, and then removed as a foil. For a Ni-alloy replica surface investigated by Plonka et al. [49] the surface roughness was less than 1 nm and the UCN transport losses by diffuse reflection, absorption or up-scattering were measured to be less than 10^{-3} per reflection. Together with the high neutron optical potential of $^{\text{nat}}$Ni or even ^{58}Ni ($V_{\text{F}}(^{\text{nat}}\text{Ni}) \sim 250$ neV, $V_{\text{F}}(^{58}\text{Ni}) \sim 340$ neV) [51] those guides are well suited to transport UCN over a longer distance to a multi-purpose experimental site.

Replica foils for the FRM II are produced by the company S-DH (Sputter-Dünnschichttechnik Heidelberg). They have a maximal surface area of 500 × 500 mm. To obtain guides, the foils have to be rolled and then welded along the longitudinal seam. Not to damage the inner surface, it is important not to weld through the natural-nickel supporting layer. If the UCN reflecting layer is touched, this can lead to uncertain effects on the inner surface, where neutrons can be lost. Currently the S-DH company is able to produce guide elements out of several pieces which are welded together to a maximum length of 3 m, which can be connected via flanges to longer guide sections. Figure 5 shows a complete replica guide with supporting rings, flanges and its cladding tube. The inner diameter of the replica tube is 115 mm, the outer diameter is 116 mm. The inner diameter of the cladding tube is 168 mm.

These guides were experimentally characterized [2,8,20,29]. Thereby the transmission and storage properties have been investigated. The transmission per unit of guide length was measured to be $T = (0.990 \pm 0.006)\ \text{m}^{-1}$ for UCN in the energy range of (100–200) neV, and by a UCN storage experiment the loss probability per wall bounce was determinded to be in the range of $(1.4\text{–}2.5) \cdot 10^{-4}$. This means that even after 40 m of UCN transport more then 50% of the initial neutron flux can reach a connected experiment.

5. Current status of the FRM II UCN source

The nuclear licensing procedures of the FRM II UCN source require a complete non-nuclear test of all components before their installation and commissioning at the FRM II. Such a test setup of all important components of the UCN source has been installed at the Maier-Leibnitz-Laboratory (MLL), see Fig. 6.

Three vessels, one filled with liquid nitrogen ($V = 12\ \text{m}^3$) and two filled with gaseous helium ($V = 15\ \text{m}^3$ each, $p = 16$ bar), have been set up outside south of the MLL building. A 70 m^2 wide and 3.7 m high hall, made out of wood, houses the two compressors (electrical power 250 kW each, $p_{\text{max}} = 14.5$ bar, helium mass flow 85 g/s) of each cold box, accompanied by two oil separators and water filled circuits for the cooling of the compressors. A small cooling tower next to the compressor hall removes the produced heat from the water cooling circuits. The helium cooling machines themselves are located inside the MLL. Two conventional cooling machines with a power of 500 W at 5 K each remove the heat from a closed cooling cycle of supercritical helium. In each machine

Fig. 6. **Left:** at the south side of the MLL building a 70 m^2 wide and 3.7 m high housing for two helium compressors and their oil removal system has been build. The two vessels on the left are filled with gaseous helium, the vessel on the right contains liquid nitrogen. **Right:** two conventional cooling machines (right hand side of the picture) produce liquid helium, which is supplied to a third cold box to remove the heat from a closed cooling loop with supercritical helium. A 1 : 1 rebuild of the beam tube SR6 (left hand side of the picture), together with all inpile parts of the UCN source, has been set up.

the compressed helium gas is precooled by liquid nitrogen to 80 K, and then further cooled down by two Brayton cycles driven by turbines to 25 K. Below this temperature two Joule-Thomson expansion valves produce liquid helium (liquefaction rate 70 l/h each) and deliver it to a third cold box. The liquid helium is then used to cool a closed loop of supercritical helium ($p = 3.4$ bar, helium mass flow 120 g/s) via two heat exchangers. This closed loop, which is driven by a special pump, is connected with cryogenic transfer lines to the converter vessel inside of the SR6 beam tube, housing the solid deuterium and the solid hydrogen.

A 1 : 1 rebuild of the beam tube SR6, where all the converter parts of the UCN source are located, has been set up. For non-nuclear tests the heat input of the FRM II to the converter is simulated by several heating devices. With all these systems in place, the UCN source has started its non-nuclear test phase. In these tests all parameters influencing the operation of the cooling machines and all necessary auxiliary systems will be varied to optimize the procedure to freeze out deuterium and hydrogen, with the simulated nuclear heat load of the FRM II. After the tests the whole cooling machine, together with the helium and liquid nitrogen vessels, will be transferred to the FRM II, and all the other parts of the source and auxiliary systems will be built and installed.

6. Possible impacts for a UCN source at the ESS

At the ESS the development and installation of VCN and UCN sources is planned within the HighNESS project. There are two principle options for a UCN source, an inpile source or a beam source, hereby meaning that an inpile location is inside the ESS monolith ($R < 5.5$ m) [21,22], and that a beam source can be placed either outside of the monolith but inside the bunker or outside of the bunker ($R > 15$ m). For inpile UCN sources four possible locations inside the monolith and two different converter materials – solid D_2 and superfluid He (He-II) – have been identified. The main challenges in all positions are the cooling of a source, the extraction of the UCN to connected experiments and the placement of all necessary equipment for cooling and extraction.

At all inpile positions the heat input to the converter material and the supporting structures will be significant. Without knowing an exact position or geometry of a future UCN source, a comparison between the FRM II liquid D_2 cold source and the ESS lower liquid D_2 moderator, which will possibly deliver cold neutrons to a future UCN source, can give an estimation of the necessary cooling power. At the FRM II the heat load on the cold source is 7 kW (26 l of liquid D_2 at 25 K), which is removed by a cooling system delivering 200 g/s of helium gas at a

temperature of 19 K [44]. At the ESS the expected heat load on the lower cold source is 57 kW (34 l of liquid D_2), which shall be removed by a mass flow of helium gas of 3.4 kg/s at a temperature of 22.5 K [9]. Comparing this and the neutron fluxes at FRM II and ESS one can expect that the heat load on a UCN source at ESS will be approximately one order of magnitude higher than on the UCN source at FRM II if the source is placed at a similar position. This means that at the ESS for a sD_2 UCN source a heat load of at least several kW at 5 K can be expected, taking into account that the heat load on the FRM II UCN source is ∼500 W. A cooling power of several kW at 5 K is technically feasible, but already requires cooling machines at the commercially available upper power limit. Concerning the fact, that for a He-II source temperatures below 1 K are desired, thus requiring even more high-power cooling techniques, a sD_2 source for the inpile case seems to be more realistic and easier achievable, although Serebrov et al. have demonstrated at the WWR-M reactor at PNPI that, with an efficient thermal shielding and powerful cooling techniques, it is possible to remove 60 W of heat from a He-II converter at a temperature of 1.37 K [55]. An effective sD_2 converter thickness of a few cm would be sufficient, if the UCN source is placed at a position where a strong cold neutron flux from the lower liquid D_2 moderator hits the deuterium converter. Also a real thin film source as described in ref. [70] could be installed at an inpile position, if a large effective converter area is available.

Considering the case of sD_2 as converter material for a future UCN source at ESS, the freeze-out procedures tested at the FRM II could be used to produce transparent crystals. Freeze-out is possible via resublimation under conditions as described in Section 3.2, or from the liquid phase. In the latter case, direct horizontal extraction of the UCN is only possible through a window, or by vertical extraction upwards followed by a 90° reflection to a horizontal direction. In any case one can expect a certain decrease of the transparency of the crystal and the formation of frost on the surface of the sD_2, as described in ref. [5]. From time to time hence a thermal cycling ("conditioning") of the crystal could be necessary to increase its transparency and remove the frost from the surface, which should be foreseen when designing the cooling facility of the UCN source.

Due to the strong irradiation of a sD_2 converter to be expected at most suitable inpile positions, the $J = 0$ population will be larger than 99% if the temperature is below 9 K, as described in Section 3.3. Although the $J = 0$ state will be fully populated after a few hours of irradiation, an initial para-to-ortho conversion of the deuterium will most probably be necessary. The thermal conductivity of ortho-deuterium at 5 K is approximately one order of magnitude higher than of deuterium with the natural para-ortho-mixture [26,27], which would strongly decrease the heat removal from the crystal. If one considers a UCN source with a small amount of deuterium as at the FRM II source, only a small and compact para-to-ortho conversion device as described in Section 3.1 would be necessary. One has to mention that meanwhile the catalyst OXISORB® is no longer produced, but most probably also other materials with a strong magnetic moment are suitable for catalytic para-to-ortho conversion, a field which is subject of current and future research.

At any inpile position the efficient transport of UCN from the source to an experiment is important, as in this case a distance of several tens of meters has to be passed. The newest generation of replica guides, as described in Section 4, are a valuable option for this purpose. With a transmission probability of 99%/m such distances can be passed with losses of less than 50%. Furthermore, these replica guides are radiation resistant, which is of particular importance at inpile positions of a UCN source at the ESS. Also adapted diameters of replica guides are possible, as these are fabricated by welding of foils of variable size.

In summary, a small sD_2 UCN source – similar as planned at the FRM II – seams a feasible option if the converter shall be placed at an inpile position close to the lower moderator. The latter already delivers neutrons in a suitable energy range, so that no further premoderation with hydrogen or deuterium is necessary. The necessary small amount of sD_2 can be cooled with commercially available helium refrigerators or liquefiers. The results of research performed during the last years at the FRM II on converter preparation, para-to-ortho conversion, radiation effects and UCN transport, can at least partially be transferred and used as input for the design and construction of a future UCN source at ESS.

Acknowledgements

The author thankfully acknowledges the support of innumerable scientists, engineers, technicians and students from the Physics Department E18 of TUM, the Maier-Leibnitz-Laboratory of TUM and LMU, the TRIGA Mainz, the ILL, the University of Applied Sciences Augsburg, the FRM II and the MLZ.

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