DOE F 1332.16
(10-90)
All other editions
are obsolete

OMB Control No.
1910-1400
Burden Disclosure
Statement on back

U.S. DEPARTMENT OF ENERGY

UNIVERSITY CONTRACTOR, GRANTEE, AND COOPERATIVE AGREEMENT RECOMMENDATIONS FOR ANNOUNCEMENT AND DISTRIBUTION OF DOCUMENTS

(See Instructions on Reverse Side)

1. DOE Report No.	**2. DOE Contract No.**	**3. DOE Funding Office**	**4. OSTI UC Category No.**
DOE/ER/45970-10	DE-FG02-02ER45970	Materials Sciences	

5. Title

Physics with ultracold and thermal neutron beams: Testing and possible application of 'low temperature Fomblin' in a neutron lifetime experiment

6. Type of Document ("x" one). Two completely legible reproducible copies should be transmitted.

a. Scientific and technical report: ☐ monthly ☐ quarterly ☐ annual ☒ final ☐ topical ☐ other

b. Conference paper: Name of conference (no abbreviations) ____________

Location (city/st/ctry) ____________

Date (mo/day/yr) ____________ Sponsor ____________

Contents: ☐ proceedings ☐ viewgraphs ☐ paper ☐ poster sessions

c. Computer Media:

Document is accompanied by ☐ magnetic tape(s) ____________

☐ diskette(s) ____________

☐ other (specify) ____________

d. Other (e.g., journal article, manuscript, manual, etc.) (specify) ____________

7. Patent Information

a. Does this information product disclose any new equipment, process, or material? ☒ No ☐ Yes If so, identify page nos.

b. Has an invention disclosure been submitted to DOE covering any aspect of this information product? ☒ No ☐ Yes
If so, identify the DOE (or other) disclosure number and to whom the disclosure was submitted.

c. Are there any patent-related objections to the release of this information product? ☒ No ☐ Yes If so, state these objections.

d. Does this material contain Proprietary information? ☒ No ☐ Yes

8. Additional information, remarks, and special handling instructions. (Continue on separate sheet, if necessary.)

9. Submitted by (Name and Position) (Please print or type)		**Phone**
Albert Steyerl, Prof. of Physics		401-874-2204
Organization	**Signature**	**Date**
University of Rhode Island	A. Steyerl	8/10/2004

August, 2004

Physics with Ultracold and Thermal Neutron Beams: Testing and possible application of 'Low Temperature Fomblin' in a neutron lifetime experiment

(Project DE-FG02-02ER45970)

Final Report DOE/ER/45970-10 for the period 2001-August, 2004
A. Steyerl, University of Rhode Island

1. **Introduction**

This project has been focused on a measurement of the mean lifetime τ_n of the free neutron with a precision better than 0.1%. The neutron β-decay

$$n \rightarrow p + e^- + \overline{\nu}_e + 783keV$$

into a proton, electron and electron antineutrino is the prototype semi-leptonic weak decay, involving both leptons and hadrons in the first generation of elementary particles. Within the standard *V-A* theory of weak interaction, it is governed by only two constants: the vector coupling constant g_V, and axial vector constant g_A. The neutron lifetime has been measured many times over decades, and the present (2004) world-average, τ_n=885.7±0.8 s [1,2], has a weighted error of ~0.1% while individual uncertainties are typically 2-10 seconds for high precision data. The highest precision claimed by an individual measurement is ~0.15%.

An improvement is required to resolve issues of the Standard Model of the electro-weak interaction as well as of astrophysics and of Big Bang theories. The focus in astrophysics is the solar neutrino deficit problem, which requires a precise value of g_A [3]. Big Bang theories require a precise τ_n-value to understand the primordial He/H ratio [4]. The strong interest of particle physicists in τ_n [5] is mainly based on a possible difficulty with the Cabibbo Kobayashi Maskawa (CKM) matrix, which describes the mixing of quark mass states by the weak interaction. Nuclear, neutron, and pion decay data, probing the mixing amplitude V_{ud} within the first quark generation, in combination with *K* and *B* meson decay data, which probe the second and third generation (V_{us} and V_{ub}), indicate a departure from the unitarity demanded by all gauge-invariant theories. The deviation of the first-row sum $|V_{ud}|^2 + |V_{us}|^2 + |V_{ub}|^2$ from unity is on the 2.3 sigma level. Including a new value for V_{us} [6] would remove the discrepancy; but the authors of [6] note an inconsistency requiring clarification.

The largest contribution to this sum is $|V_{ud}|^2$ which is determined most sensitively by the neutron lifetime and the neutron decay asymmetry parameter *A*. Confirmation of non-unitarity would imply that the Standard Model of particle physics may have to be extended.

To prepare for an improved τ_n measurement based on ultracold neutron (UCN) storage our project had two main goals:

(a) To investigate the suitability of a new type of per-fluorinated oil for low-loss wall coating. Like Fomblin oil, which has been used in several previous high-precision τ_n measurements [7-10], the new oil consists only of carbon, oxygen and fluorine. These elements have very low neutron absorption cross sections. However, due to weak intermolecular binding the new polymer solidifies at a lower temperature (~150 K vs. ~230 K for Fomblin) and can, therefore, be used in liquid form at a lower temperature. This is important since a liquid perfectly seals small gaps and the low temperature ensures that the loss due to thermal-inelastic and quasi-elastic scattering is also small. The new types of oil have become known as 'Low Temperature Fomblin' (LTF).

(b) If indeed the anticipated low losses were obtained we planned to perform first direct UCN storage experiments in a gravitational storage system coated with this oil. This system in principle allows measurement of the storage lifetime as a function of UCN energy and trap size, and an extrapolation to zero loss yields the neutron lifetime [11].

2. **Quasi-elastic and inelastic UCN scattering studies**

For ultracold neutrons stored in low-loss traps the dominant loss is due to β-decay, the quantity to be studied. Additional losses are typically on the percent level and caused by imperfect wall reflection, scattering on the residual gas and, possibly, gaps at mechanical shutters or due to incomplete wall coating with the reflector film. Reflection losses occur because in total reflection the neutron wave penetrates a depth of ~20 nm into the wall, making it susceptible to nuclear capture, inelastic scattering to thermal energies, or quasi-elastic scattering. The latter process leads to loss if the UCN gains energy sufficient for the scattered UCN energy to exceed the wall scattering potential, however slightly this may be.

In dedicated studies of various types of LTF we measured the dependence of quasi-elastic up-scattering on energy transfer and temperature, comparing the data to ordinary Fomblin grease and oil at room temperature. At T<180K the two types of LTF used showed >3 times lower quasi-elastic scattering probabilities than Fomblin, thus satisfying one requirement for an improved τ_n measurement. Furthermore, comparing our *up-scattering* data for Fomblin grease at room temperature to our earlier quasi-elastic *down-scattering* data we showed that there is about as much scattering with a slight energy gain as with energy loss. Plausible models of quasi-elastic scattering were discussed and the results have been summarized in [12-15]. The findings indicate that earlier reports of "slight heating" or of "slight cooling" of UCN by various groups showed only "one side of the coin", i.e. up- and down-scattering are only different aspects of the same process: quasi-elastic scattering. The new results can also explain some of the "UCN anomalies" reported earlier [16].

Sufficient reduction of quasi-elastic scattering by the wall coating is one requirement for low-loss UCN storage for measuring τ_n. Thermal inelastic scattering also had to be shown to be sufficiently weak in the range T<170K anticipated for the lifetime measurements. The combination of all losses, including absorption, is conveniently expressed by a reflection loss coefficient $\eta(T)$. Temperature dependent measurements of cold neutron inelastic scattering cross sections for various LTFs were performed at Dubna at wavelength $\lambda = 1 – 2$ nm. They indicated that a factor 5 improvement in $\eta(T)$ can be expected at ~200K in comparison with room temperature Fomblin oil [17] which had previously been used in the most sensitive lifetime experiments performed so far. The gain factor at ~120 K should exceed a factor 10. Transmission experiments with very cold neutrons (velocity 6 – 15 m/s) at the ILL suggested similar improvement factors [18]. Low wall losses are a critical issue since a determination of τ_n from storage data always requires an extrapolation to zero wall loss, and any extrapolation process might be expected to become more reliable as all non-decay losses are reduced.

In light of our first experimental data, discussed below, the preceding statement can be made more specific. We have indeed achieved the lowest wall reflection loss coefficient ever reported in UCN storage in material traps. Nevertheless, the task of reliable extrapolation of measured storage lifetimes to obtain τ_n has turned out to be very difficult. This is a common problem of all schemes of neutron lifetime measurement, including magnetic storage where loss due to depolarization cannot be excluded *a priori*. We are planning to take these concerns into account by use of an improved UCN storage system.

3. **First UCN storage data with an LTF coated gravitational trap**

In the experiments performed during two reactor cycles at the ILL reactor we used the gravitational UCN storage system shown in Fig. 1, whose revolving cylindrical trap has an opening on the periphery. In the emptying phase, the trap is rotated intermittently such that the UCN “poured off” at each step are only a “high-energy” fraction of the UCN remaining in the trap after the preceding steps. A typical time diagram of a measurement cycle is shown in Fig. 2. For measurements with different storage times t_{st}, we measured the integral count-rate for each of the five counting periods (the intervals *5* to *9* in Fig. 2), to determine the storage lifetime as a function of UCN energy and temperature. Calculation of parameters like the mean energy E and wall collision frequency f for each counting interval requires calculations based on assumptions about the UCN spectrum in the trap and, most importantly, based on a model for the wall potential.

Using the elementary theory of the E-dependence of UCN reflection loss for a semi-infinite uniform wall potential, each energy band is characterized by γ, the mean wall collision frequency for the UCN counted in this spectral range, weighted by the reflection loss probability. For an *ideal step function* potential, elementary theory predicts the relationship

$$\tau_{st}^{-1} = \tau_n^{-1} + \eta\gamma, \qquad (1)$$

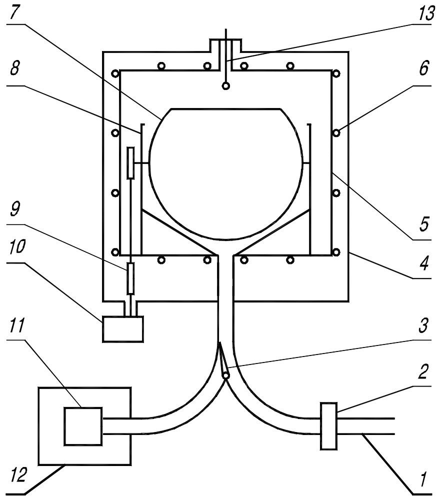


Fig. 1: Scheme of the gravitational system. *1* – UCN guide; *2* – intake valve; *3* – steering valve; *4* – external vacuum (~5×10^{-5} mbar); *5* – "clean" vacuum (~1×10^{-6} mbar); *6* – cooling coils; *7* – UCN trap; *8* – N_2-cryostat; *9* – mechanics for trap rotation; *11* – UCN detector; *13* – oil evaporator.

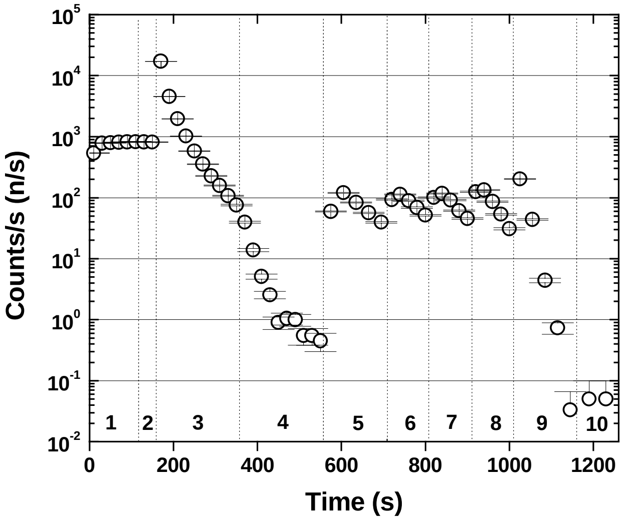


Fig. 2: Example of measuring cycle. In a cyclic way, a short storage time (interval 4), e.g., 200s, alternates with a long time, e.g., 1200s, leaving all the other parameters the same. Combination of the data allows us to determine the storage lifetime τ_{st} as a function of temperature and UCN energy. Five energy intervals are used (the counting regions 5 – 9).

where $\eta = -\mathrm{Im}(V_F)/\mathrm{Re}(V_F)$ is a constant determined by the real and imaginary parts of the mean Fermi scattering potential for the wall material at a given temperature. For the type 'LTF2' used ($CF_3O(CF_2O)_n(CF_2CF_2O)_m(OCF_2CF_2O)_kCF_3$ with n = 30.3, m = 1.5, k = 0.2), $\mathrm{Re}(V_F) \approx 120$ neV at a temperature of -120ºC. Since (1) is linear in γ, plotting τ_{st}^{-1} *vs* γ gives a straight line, and the neutron lifetime τ_n is obtained as its extrapolation to $\gamma = 0$, the limit of zero wall loss.

In our first experiments we used a Ti-coated Cu trap with diameter 76 cm and width 50 cm. The opening extended over a large angle of 64º to enhance the UCN efflux speed. Ti was chosen to allow a stringent test of the quality of the oil coating on top of Ti. Since Ti has a negative scattering length, gaps and spots where the oil film on the titanium is too thin would drastically reduce the storage lifetime.

The oil was sputtered in vacuum onto the cold inner trap surface from a heated oil reservoir, which was moved to different positions inside the trap to ensure uniform wall coverage. After several evaporations, a mean storage lifetime of $\langle\tau_{st}\rangle = 867\pm2$ s (averaged over all UCN energies in the trap) was achieved in the temperature range from -150 to -160ºC. This value is only 19 s, or 2%, away from $\tau_n \approx 885$ s. The small wall reflection loss rate translates into a loss coefficient $\eta = 5 \times 10^{-6}$. For comparison, the lowest η value

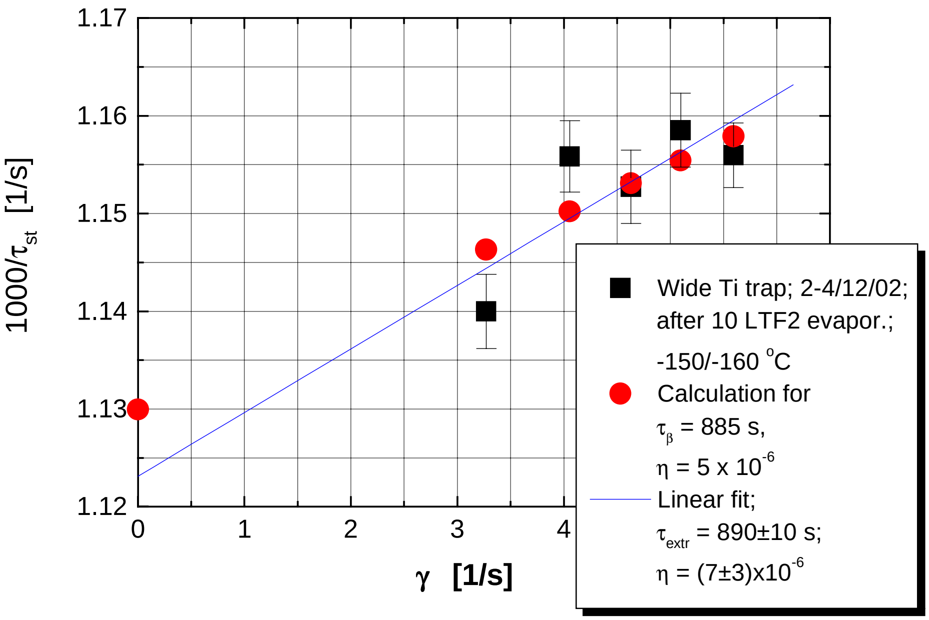


Fig. 3: Preliminary plot of inverse storage lifetime data vs. γ for a two-day run with a wide LTF2 covered Ti trap at –150 to -160ºC. If the oil film can be represented by an ideal step function potential for the UCN, the straight-line extrapolation to $\gamma = 0$ intersects the *y*-axis at τ_n^{-1}. The circular dots are a calculation for $\tau_n = 885$ s and $\eta = 5 \times 10^{-6}$.

reported previously for any material had been (6.1±0.6) x 10^{-6} for solid oxygen at the much lower temperature of 13 K [11].

To illustrate the extrapolation procedure, Fig. 3 shows the inverse storage lifetime (the square dots) plotted vs. γ for a measurement period of two days.

Additional measurements with a Be-coated trap were carried out to search for a possible difference with the Ti-trap. If the oil film did not completely cover the surface, the Be would still be a good reflector at the spots with insufficient coating, while uncovered Ti does not reflect UCN. However, the storage lifetime measured with Be, $\langle\tau_{st}\rangle = 872 \pm 1.5$s, corresponded to the same reflection loss coefficient $\eta \approx 5\times10^{-6}$ as for the Ti substrate, the difference being solely due to the larger size and different shape of the Be-trap. This indicates that, on either trap, reproducible oil films have been obtained, independently of the substrate material. For the Be trap a clear deterioration of $\tau_{st}(E)$ by a few seconds occurred within a few days. This indicates a gradual contamination from the rest gas at ~10^{-5} mbar. It is likely that a similar short-term degradation occurred also for the Ti-trap.

While the η-value was low it was still ~10 times higher than had been expected for LTF at ~110 K [17]. Moreover, a serious uncertainty of interpretation arises if we cannot be sure that the elementary theory of UCN reflection from a thick wall with uniform scattering potential applies. This happens in the presence of impurities on the wall or other deficiencies of wall coating, especially if these are not stable in time. In such cases, (1)

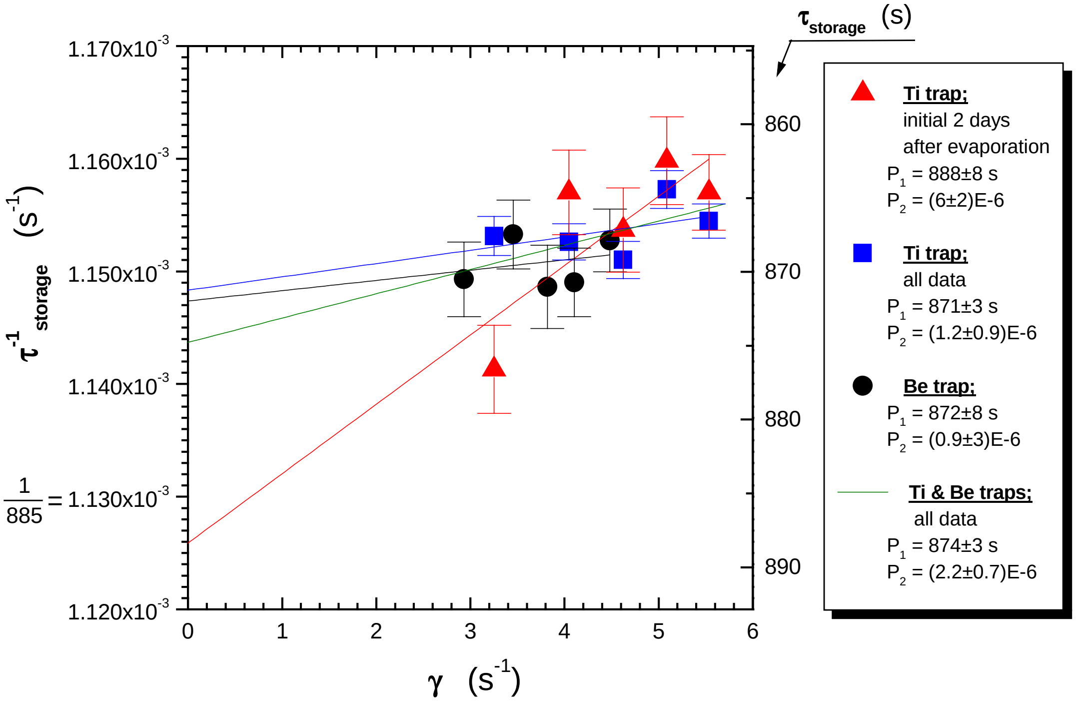


Fig. 4: An extrapolation to $\gamma = 0$ for initial storage lifetime data with a Ti trap (the first two days after evaporation) is compared to extrapolations for long-time data (~10 days) with Ti and Be traps. A combination of these data is also shown. The large variation of extrapolation results is also observed for a different data subset combining one week's long-time data for the same Be trap: $P_1 = 882.5 \pm 4.7$ s (= extrapolation to γ=0), $P_2 = (5.3 \pm 1.8) \times 10^{-6}$ (= slope of τ_{st}^{-1} *vs.* γ).

will not be exact and the straight-line fit and extrapolation are inadequate. Fig. 4 shows an example where quite different extrapolation results were obtained for three different data sets. An extrapolation for all the data is also included.

A combination with data for traps with different shapes and sizes, e.g. a narrow and a wide cylinder, can help to extend the γ-range covered by the data. However, we must be sure that the quality of oil coating is always the same, as well as that the model of the same step-function potential applies. If it does not, size extrapolation is not applicable, either, since for the gravitational spectrometer used the dependency on the ideal wall condition is not relaxed by size variation.

More details of these measurements and the data analysis were presented in several reports [19-22] and ILL beam time applications [23,24].

4. **Project modification**

These uncertainties of data interpretation and divergent views of remedial strategies and presentation of preliminary results were at the heart of a decision to adopt a modification of the 'LTF project'. For the new plans, a group from the Joint Institute for Nuclear

Research, Dubna, has taken the place of the group from the Petersburg Nuclear Physics Institute, Gatchina, the other collaborating Institutes remaining the same.

The difficulties with the gravitational system used can be summarized as follows: The measurements with the gravitational system provide the UCN energy-dependence of storage lifetime, and this dependence on energy (and on trap size) can be used to extrapolate to zero collision frequency, i.e. no wall loss. However, this extrapolation to the neutron lifetime τ_n turned out to be unreliable. The uncertainty resulted (a) from a possible deviation from the linear extrapolation, which is valid only for UCN interaction with an ideal step-function potential representing the wall; and (b) the requirement of a detailed knowledge of the UCN spectrum for all modes of operation of the system. Our experiments showed that neither condition (a) nor (b) could be satisfied. The wall potential is not ideal but modified by contamination, probably from the residual gas. This was visible in a deterioration of measured storage lifetime by several seconds within a few hours up to days after refreshing the oil coating. Moreover, no "reasonable" model for the spectral development during a measurement cycle was found. The difference between measured and calculated relative intensities of counts in the counting periods 5-9 of Fig. 2 was up to 25%. Doppler shifts due to trap rotation were one possibility for uncontrollable spectral changes. It has become obvious to us that these intrinsic problems required development of an improved UCN trap.

Rather than relying on the UCN energy dependence of reflectivity we returned to the principle of trap size variation, which had been applied successfully for the first time in Ref. [7]. This scheme allows us to use a special scaling technique to achieve a high degree of independence from the details of, both the UCN-wall interaction and the UCN spectrum. It also allows us to vary the trap size in a range much wider than possible in the gravitational system, thus making extrapolation more precise. Most importantly, the system ensures that the extrapolation to the neutron lifetime is reliable. Finally, the possibility to use a broad UCN spectrum translates into high UCN intensity and good statistics.

The design of the new system had to be consistent with the following special requirement for the LTF oil: In contrast to ordinary Fomblin oil, the new LTF oil is difficult to produce and therefore available only in small quantities. Hence, to change the storage volume in a wide range we cannot use a design based on a wall moveable like a piston inside a rectangular volume, as in [7,10]. Such a system requires a continuous ample supply of the oil to fill the gap at the edges of the moveable wall. To avoid gaps when the storage volume is changed, we use an 'accordion-like system', i.e. a highly flexible bellows. This modification will allow us to take data not only for liquid oil, but also down to ~100 K, well below the solidification temperature of ~150 K. The expected wall reflection losses are even smaller in this range.

The new system has been described in detail in [25,26]. The most prominent advantage of using a bellows is as follows: The total surface area and its distribution of surface area over height remain constant while the volume is changeable in a wide range. This unique characteristic, in combination with application of the scaling technique, ensures that a plot

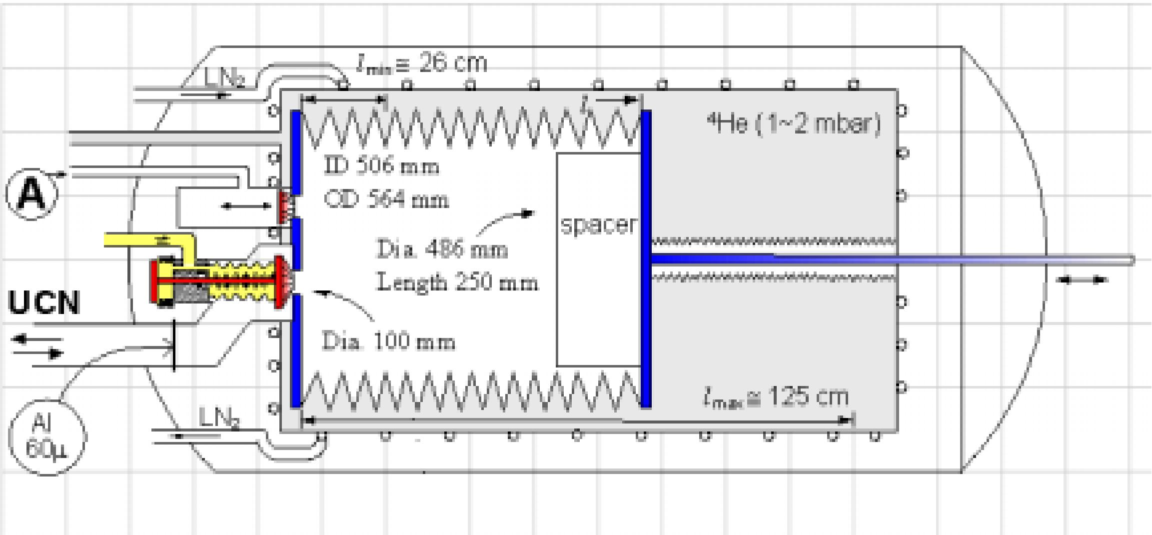


Fig. 5: The main feature of the new UCN storage system using 'low-temperature Fomblin' is a flexible bellows confining the UCN in a trap whose volume is variable while the surface area is constant. As in an accordion, the size changes without the danger of gaps opening up. Inserting a spacer into the trap volume, as shown, allows an extension of the dynamical range, i.e. a wide range of wall collision frequency can be covered.

of measured inverse storage lifetimes versus collision rate (or efflux speed at the end of storage) is almost exactly linear and intersects the *y*-axis at the inverse neutron lifetime. This is true under gravity and for any UCN spectrum and for any energy dependence of wall losses, which may also include leakage through small gaps. An important condition has already been noted in [7]: the UCN spectrum should be cut from below and contain no energies insufficient for the neutrons to reach the top of the trap. As long as this condition is satisfied the results of calculations are highly independent of the initial UCN spectrum entering the trap.

Fig. 5 shows how the bellows is fitted into a horizontal, cylindrical vacuum chamber. We had used the same vacuum vessel in a previous experiment, and this equipment, including pumps, valves and measuring gauges, is available. The same is true for the LTF oil and the motors and control units needed to change the trap volume with the required precision of <0.01mm.

Fig. 6 shows a calculation of inverse storage lifetime *versus* wall collision rate [25,26]. Linear fits for different subgroups of points for volumes in the range $V_{max}/V = 1$ to 27 give τ_n extrapolations within 0.5 s of the initial input value 886 s. Even the four low-volume points farthest from the *y*-intersection (window on right) show no deviation exceeding the anticipated accuracy of the proposed τ_n experiment. Using a wall model with a velocity dependence of losses very different from the step function model, and

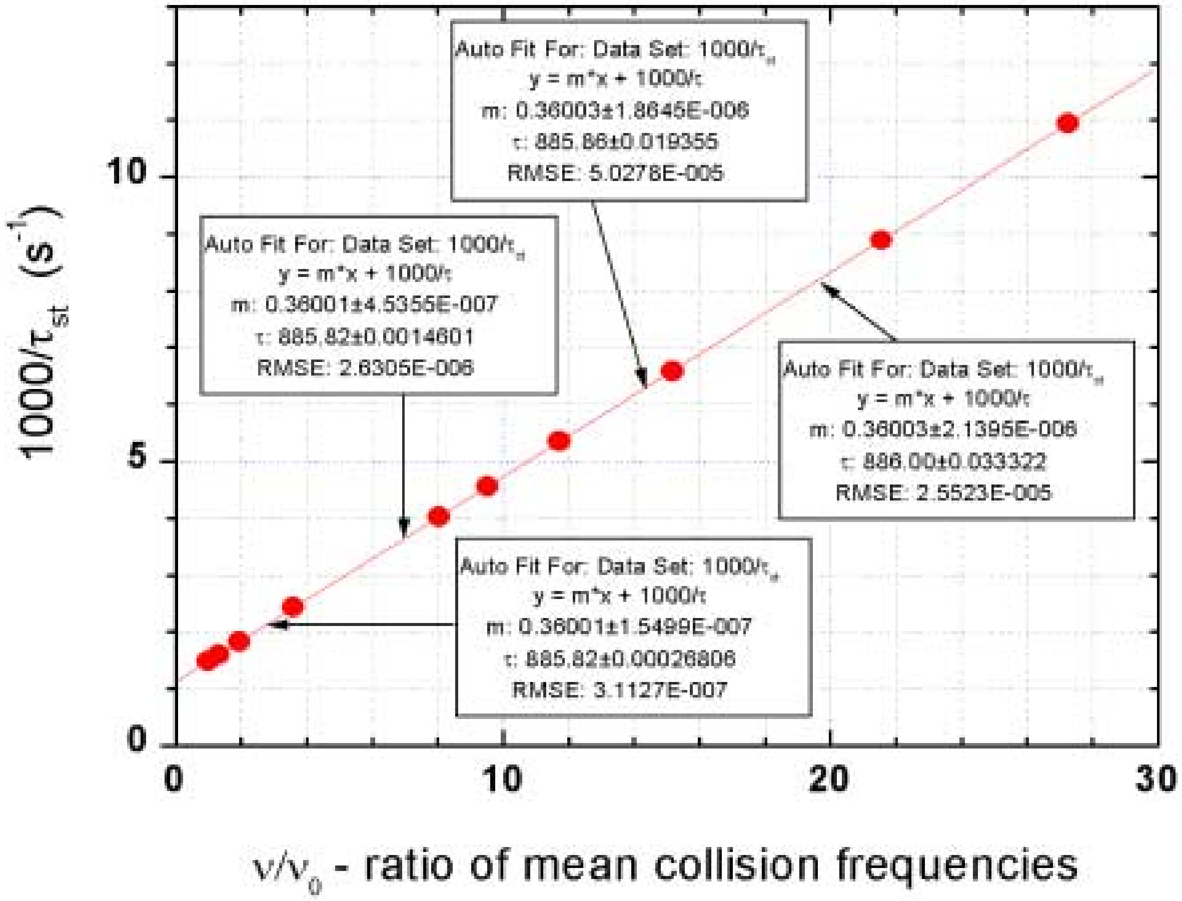


Fig. 6: Test of linearity of a plot of inverse storage lifetime, τ_{st}^{-1}, versus relative wall collision rate ν/ν_0. For the largest trap volume V_{max}, $\nu_0 = 51.9$ Hz. The τ_{st}^{-1}-values are mean values for the storage interval from $t_1 = 300$ s to $t_2 = 2300$ s (for the largest volume). The collision rates are mean values for the same time interval. The windows show results of linear fits for subgroups of points (counting from the left): bottom: 1-4; left: 1-5; top: 1-10; right: 7-10. The extrapolated y-axis intersections agree within 0.3 s with the value $\tau_n = 886$ s used in all calculations. The error ranges reflect residuals and do not include counting statistics.

adding gaps, we obtained the same degree of linearity. The same degree of linearity is also obtained if we plot against efflux rate instead of collision rate. This plot has the advantage that the efflux rate is directly measurable whereas the collision rate requires calculation.

Further special features of the new system are:

(a) The UCN shutter design ensures a high degree of leak-tightness by precise manufacturing and application of pressure to the edge-shaped contact area between the static and moving parts.

(b) We can check for a residual small penetration-rate by measuring the leakage of helium gas through the closed gate.

(c) The LTF oil will be deposited on the trap walls by condensation from the vapor, which is admitted into the storage space from a heated reservoir *A* (Fig. 5). The oil vapor is transported in ~10 mbar of helium-4. This method had previously been used with D_2O vapor and had produced a wall coating with a uniform layer of a thickness variable with 0.1-0.2 nm precision.

(d) Since the wall losses are temperature dependent we need good temperature uniformity over the entire trap surface. This is achieved by surrounding the trap completely with a radiation shield, which is kept at a constant temperature variable between ~100K and room temperature. A good thermal contact between the shield and the storage chamber is provided by 1-2 mbar of helium gas.

(e) The helium-filled space surrounding the storage volume also serves as a buffer zone, reducing the risk of oil contamination by hydrogen and nitrogen in the residual gas of the trap.

5. Present status

The crucial element of the new system is the large highly flexible welded bellows with a mean diameter of ~56 cm and a length of 125 cm (stretched) and 26 cm (compressed). It has been purchased and installed in the vacuum tank in the workshop at the Frank Laboratory of the Joint Institute for Nuclear Research, Dubna. The entire system is scheduled to be completed and shipped to the ILL in September, 2004. Due to refurbishment work the ILL reactor will not operate in the Fall of 2004, but we hope to obtain beam-time in 2005. P. Geltenbort will coordinate this work with the requirements of the UCN facility at the ILL.

6. Personnel associated with the project

Graduate Student O. Kwon is fully integrated into this project and he has obtained support under this project for a Research Assistantship during the academic year and the summer.

References

1 B.G. Yerozolimsky, Nucl. Instr. Meth. Phys. Res. A **440** (2000) 491; H. Abele, Nucl. Instr. Meth. Phys. Res. A **440** (2000) 499.
2 K. Hagiwara *et al.* (Particle Data Group), *Review of Particle Physics*, Phys. Rev. D **66** (2002) 765; and 2003 partial update at http://pdg.lbl.gov.
3 A. Bertin, M. Capponi *et al.*, Phys. Lett. B 199 (1987) 108.
4 R.E. Lopez, M.S. Turner, Phys. Rev. D **59** (1999) 103502-1.
5 I.S. Towner, J.C. Hardy, in *Physics beyond the Standard Model* (World Scientific 1999).
6 A. Sher, R. Appel *et al.*, Phys. Rev. Lett. **91** (2003) 261802.
7 W. Mampe *et al.*, Phys. Rev. Lett. **63** (1989) 593.

8 Mampe *et al.*, Sov. Phys. JETP **57** (1993) 82.

9 S. Arzumanov *et al.*, in ISINN-5, *Neutron Spectroscopy, Nuclear Structure, Related Topics*, Dubna, E3-97-213, 1997, p. 53.

10 A. Pichlmaier *et al.*, Nucl. Instr, Meth. Phys. Res. A **440** (2000) 517.

11 V. Nesvizhevsky *et al.*, Sov. Phys. JETP **75** (1992) 405.

12 P. Geltenbort, A. Steyerl, B. Yerozolimsky et al., *Quasi-elastic spectral change of stored UCN*, ISINN-9, *Neutron Spectroscopy, Nuclear Structure, Related Topics*, Joint Institute for Nuclear Research, Dubna, E3-2001-192, 2001, p. 415.

13 A. Steyerl, B.G. Yerozolimsky, A.P. Serebrov et al., *Experimental study of quasi-elastic scattering of ultracold neutrons*, Eur. Phys. J. B **28** (2002) 299; document DOE/ER/45970-03.

14 A. Steyerl, B. Yerozolimsky et al., *Quasi-elastic UCN scattering*, Internat. Seminar on Interaction of Neutrons with Nuclei, ISINN-10, in *Neutron Spectroscopy, Nuclear Structure, Related Topic*, Joint Institute for Nuclear Research, Dubna, E3-2003-10, p. 388-396; document DOE/ER/45970-04.

15 A. Steyerl, B. Yerozolimsky, A. Serebrov et al., *Quasi-elastic wall reflections of ultracold neutrons*, Amer. Conf. Neutr. Scatt. (ACNS), June 2002, http://www.sns.gov/acns.

16 V. P. Alfimenko, A. V. Strelkov et al., Sov. Phys. JETP Lett. **55** (1992) 84.

17 Yu.N. Pokotilovski, Nucl. Instr. Meth. Phys. Res. A **425** (1999) 320; JETP **96** (2003) 172.

18 V. Morozov et al., *Measurement of total cross sections for the very cold neutron interaction with a series of materials in the temperature range 80-300K*, International Seminar on Interaction of Neutrons with Nuclei, ISINN-10, in *Neutron Spectroscopy, Nuclear Structure, Related Topic*, Joint Institute for Nuclear Research, Dubna, E3-2003-10, p. 376-381.

19 A. Steyerl *et al*., ILL Experimental Report *Preparation of neutron lifetime experiment using 'low temperature Fomblin*, Institut Laue-Langevin, Jan. 2003, "TEST-690"; document DOE/ER/45970-05.

20 P. Geltenbort, *Recent neutron lifetime experiments at the ultracold neutron source of the ILL*, International Seminar on the Interaction of Neutrons with Nuclei, ISINN-11, Joint Institute for Nuclear Research, Dubna, May 28-31, 2003.

21 P. Geltenbort *et al.* (presented by A. Steyerl), *Preparation of a new neutron lifetime experiment*, Fourth Internat. Conf. 'Ultracold and Cold Neutrons, Physics and Sources', St. Petersburg, June 16-21, 2003, http://nrd.pnpi.spb.ru/UCN_CNS/ucntalks.htm; document DOE/ER/45970-06.

22 P. Geltenbort *et al.*, *Preparation of a new neutron lifetime experiment using 'low temperature Fomblin'*, paper prepared for proceedings of ISINN-11, Joint Institute for Nuclear Research, Dubna, May 28-31, 2003; http://surface.phys.uri.edu/Publications-Steyerl/n-lifetime.2003.pdf; document DOE/ER/45970-07.

23 A. Steyerl et al., ILL Research Proposal for the proposal round 2003, April-July; document DOE/ER/45970-08.

24 A. Steyerl et al., ILL Research Proposal for the proposal round 2003, August-October; document DOE/ER/45970-09.

25 B. Yerozolimsky, A. Steyerl *et al.*, *Neutron lifetime experiment based on an 'accordion-like' UCN storage volume coated with 'Low Temperature Fomblin'*, International Conf. on Preci-

sion measurements with cold neutrons, NIST, April 5-7, 2004; to be published; document DOE/ER/45970-11.

26 B. Yerozolimsky, A. Steyerl *et al.*, *'Low Temperature Fomblin' coated accordion-like UCN storage system for neutron lifetime experiment*, International Seminar on Interaction of Neutrons with Nuclei, ISINN-12, Joint Institute for Nuclear Research, Dubna, 26-29 May, 2004; to be published; document DOE/ER/45970-12.

DOE F 1332.16
(10-90)
All other editions
are obsolete

OMB Control No
1910-1400
Burden Disclosure
Statement on bac

U.S. DEPARTMENT OF ENERGY

UNIVERSITY CONTRACTOR, GRANTEE, AND COOPERATIVE AGREEMENT RECOMMENDATIONS FOR ANNOUNCEMENT AND DISTRIBUTION OF DOCUMENTS

(See Instructions on Reverse Side)

1. DOE Report No. DOE/ER/45970-11	**2. DOE Contract No.** DE-FG02-02ER45970	**3. DOE Funding Office** Materials Sciences	**4. OSTI UC Category No.**

5. Title Neutron lifetime experiment based on an ‘accordion-like’ UCN storage volume coated with ‘Low Temperature Fomblin’, by B. Yerozolimsky, A. Steyerl, O. Kwon, V. Luschikov, E. Lychagin, A. Muzychka, A. Strelkov, P. Geltenbort, N. Achiwa, A. Pichlmaier, P. Fierlinger

6. Type of Document (“x” one). Two completely legible reproducible copies should be transmitted.

a. Scientific and technical report: ☐ monthly ☐ quarterly ☐ annual ☐ final ☐ topical ☐ other

b. Conference paper: Name of conference (no abbreviations) International Seminar on Precision Measurements with Cold Neutrons

Location (city/st/ctry) Gaithersburg, MD, U.S.A.

Date (mo/day/yr) 4/05-09/2004 Sponsor National Institute for Standards and Technology

Contents: ☐ proceedings ☐ viewgraphs ☒ paper ☐ poster sessions

for proceedings

c. Computer Media:

Document is accompanied by ☐ magnetic tape(s)

☐ diskette(s)

☐ other (specify)

d. Other (e.g., journal article, manuscript, manual, etc.) (specify)

7. Patent Information

a. Does this information product disclose any new equipment, process, or material? ☒ No ☐ Yes If so, identify page nos.

b. Has an invention disclosure been submitted to DOE covering any aspect of this information product? ☒ No ☐ Yes
If so, identify the DOE (or other) disclosure number and to whom the disclosure was submitted.

c. Are there any patent-related objections to the release of this information product? ☒ No ☐ Yes If so, state these objections.

d. Does this material contain Proprietary information? ☒ No ☐ Yes

8. Additional information, remarks, and special handling instructions. (Continue on separate sheet, if necessary.)

9. Submitted by (Name and Position) (Please print or type) Albert Steyerl, Prof. of Physics		**Phone** 401-874-2204
Organization University of Rhode Island	**Signature** A. Steyerl	**Date** 8/10/2004

NEUTRON LIFETIME EXPERIMENT BASED ON AN ‘ACCORDION-LIKE’ UCN STORAGE VOLUME COATED WITH ‘LOW TEMPERATURE FOMBLIN’

B. Yerozolimsky[1], A. Steyerl[2], O. Kwon[2], V. Luschikov[3], E. Lychagin[3], A. Muzychka[3], A. Strelkov[3], P. Geltenbort[4], N. Achiwa[5], A. Pichlmaier[6], P. Fierlinger[6]

[1] Harvard University, Cambridge, MA, USA; [2] University of Rhode Island, Kingston, RI, USA; [3] Joint Institute for Nuclear Research, Dubna, Russia; [4] Institut Laue Langevin, Grenoble, France; [5] Osaka University, Japan; [6] Paul Scherrer Institut, Villigen, Switzerland

Key words: neutron lifetime; ultracold neutrons

A new type of per-fluorinated polymer, ‘Low Temperature Fomblin’, has been tested as a wall coating in an ultracold neutron (UCN) storage experiment using a gravitational storage system. The data show a UCN reflection loss coefficient η as low as $\sim 5\times10^{-6}$ in the temperature range 105 - 150 K. We plan to use this oil in a new type of neutron lifetime measurement, where a bellows system (‘accordion’) enables to vary the trap size in a wide range while the total surface area and distribution of surface area over height remain constant. These unique characteristics, in combination with application of the scaling technique developed by W. Mampe et al. in 1989, ensure exact linearity for the extrapolation from inverse storage lifetimes to the inverse neutron lifetime. Linearity holds for any energy dependence of loss coefficient $\mu(E)$. Using the UCN source at the Institut Laue Langevin we expect to achieve a lifetime precision below ±1 s.

1. **Introduction**

Particle decay data indicate that the Cabibbo-Kobayashi-Maskawa matrix may deviate from unitarity (presently at the 2.7-sigma level [1]). This question depends critically on the up-down quark mixing amplitude V_{ud}, which is determined most sensitively by the neutron lifetime τ_{n} and the neutron decay asymmetry coefficient A. A reliable, precise value of τ_{n} will also help to refine models of astrophysics [2] and cosmology [3]. The current world average is $\tau_{\mathrm{n}} = 885.7 \pm 0.8$s [4]. We propose a new τ_{n} measurement with a precision below 1s, using UCN storage. Wall losses are minimized by the use of 'low-temperature Fomblin', and the notoriously difficult extrapolation from storage lifetimes to τ_{n} is made more reliable by the novel use of an accordion-like storage vessel. In this system, the surface area and its distribution over height remain constant while the volume is changeable in a wide range. Combination with the scaling technique of Mampe *et al.* [6] ensures that the extrapolation function becomes strictly linear for any shape of UCN spectrum in the trap and for any energy dependence of reflection loss coefficient μ. Moreover, no correction for gravity is required. These unique features distinguish this method from all previous τ_{n} experiments based on UCN storage in material traps [5-10].

2. **Basic considerations**

In UCN-storage based τ_{n} experiments it is crucial that any non-decay loss due to wall collisions, gaps and the residual gas are reliably subtracted from the total loss rate. In the elementary theory of wall reflection loss the interaction with the wall atoms is described by a step-function barrier determined by the optical (or mean Fermi) potential U–iW. For an isotropic UCN distribution the mean loss probability per bounce is given by $\mu(E)$ =

$2\eta\{(U/E)\arcsin(E/U)^{1/2} - [(U-E)/E]^{1/2}\}$, where $\eta = W/U$ and E is the neutron kinetic energy at the impact point. The wall loss probability per second is

$$\tau_w^{-1} = \langle\nu\mu(E)\rangle \qquad (1)$$

where ν is the wall collision rate for a UCN and the average is taken over the UCN spectrum and trap surface. Neglecting gravity, $\nu = \mathrm{v}/\lambda$ can be expressed by the mean UCN velocity v and the mean free path $\lambda = 4V/S$. λ is independent of v and determined only by the total trap surface S and volume V. As shown in [6] this gas-kinetic result is also valid under gravity provided the trap geometry has a horizontal plane of symmetry (as in the 'accordion-trap' discussed below) and all UCN have enough energy to reach the highest point(s) in the trap. For gravitationally bent paths we use the straightforward definition $1/\lambda$ = total wall collision rate $N\nu$ divided by the volume-integrated UCN flux Φ in the trap (see Sec. 4).

However, gravity does induce an important difference. For our trap geometry, the total UCN number N is not exactly proportional to volume V, even for identical spectra. As a consequence we will plot storage data vs. ν, not $1/\lambda$, to obtain a linear dependence.

If we are sure that a single constant (η) can be factored out of the function $\mu(E)$, Eq. (1) may also be written in the form $\tau_w^{-1} = \eta\gamma$, where γ is defined as $\gamma = \langle(\mathrm{v}/\lambda)(\mu/\eta)\rangle$, averaged over spectrum and surface. However, this is strictly justified only for a smooth, uniform wall without surface contamination and/or microstructure (cracks, roughness etc.), which can be represented by a potential step function.

In an experiment we measure the numbers $N(t_1)$, $N(t_2)$ of UCN counted after storage times t_1 and t_2. Although for a broad UCN spectrum the decay curve is non-exponential one can define a mean storage lifetime for time interval t_1, t_2 in the form

$$\tau_{st} = (t_2 - t_1)/\ln[N(t_1)/N(t_2)] \qquad (2)$$

with $\tau_{st}^{-1} = \tau_n^{-1} + \tau_w^{-1}$ + other losses.

Three different methods have been used to extract τ_n from storage experiments.

(a) In Refs. [5,6,10], the vessel geometry was changed to vary λ and extrapolate the measured dependence $\tau_{st}^{-1}(\lambda^{-1})$ to $\lambda^{-1} \rightarrow 0$.

(b) In Ref. [7], the storage lifetime was measured for different intervals of UCN energy, corresponding to different mean γ-values, and the dependence $\tau_{st}^{-1}(\gamma)$ was extrapolated to $\gamma \rightarrow 0$.

(c) In Ref. [9], storage lifetimes were measured together with the flux of UCN thermally upscattered at the trap walls. This provided a further handle on the separation of wall losses from beta-decay.

Restricting the discussion to the extrapolation methods (a) and (b), the accuracy and reliability of the *y*-axis intersection is determined by several criteria:

i) First of all, the reliability of the extrapolation law $\tau_{st}^{-1}(\lambda^{-1})$ or $\tau_{st}^{-1}(\gamma)$ (or e.g. $\tau_{st}^{-1}(\nu)$) used to bridge the gap Δ_1 from $(\lambda^{-1})_{min}$ or γ_{min} (or similar *x*-variables) to zero.

ii) The maximum *x*-range Δ_2 between $(\lambda^{-1})_{min}$ and $(\lambda^{-1})_{max}$ (or similar) accessible to the experiment. If the dependence is linear and the statistical uncertainties are constant, the precision of the fitted *y*-intersection (τ_n^{-1}) is determined by the ratio Δ_2/Δ_1, and independent of the size of the gap Δ_1 (the distance of τ_{st} from τ_n).

iii) In addition to the experimental *y*-errors the uncertainties of calculated mean *x*-values like γ must be taken into account [11]. They include spectral and model uncertainties, most critically those related to the assumption of a step-function potential for the wall.

Method (a) ensures a linear dependence *if* all τ_{st} data are obtained with the same UCN energy spectrum at equivalent times of a storage cycle, so that only λ changes when the trap geometry is changed. The linearity can be checked for the experimental data, and the reliability of this test will also improve with increasing Δ_2/Δ_1. To ensure identical spectra in storage measurements with different λ, Pendlebury, Mampe *et al.* developed a scaling procedure where all time intervals Δt are chosen proportional to λ [6]. In this case, the total number of wall collisions in equivalent time intervals is the same, and therefore the spectra and spectral changes are practically the same for cycles with different λ-values. In Ref. [6], only small corrections were needed to take into account the 'loading effect' (essentially the role of β-decay during trap loading and emptying) and gravity. Both corrections are essentially reduced for the system described below.

Method (b) relies on the validity of the assumed energy dependence of the wall loss. The calculation of γ-values is directly based on the step function potential model for the wall. Experiments show that for low-absorbing materials like per-fluorinated polymers at low temperature or solid oxygen the loss coefficient is significantly higher than calculated for a clean surface [12]. Moreover, in a $\sim 10^{-5}$ mbar vacuum at ~120 K a clear deterioration of τ_{st} by several s was observed over time periods of hours to days [11], possibly due to surface contamination from the residual gas. Besides, calculation of the γ-values required for the energy extrapolation method depends on the UCN spectra. Therefore all spectral changes over a cycle must be known, including Doppler shifts due to trap rotation, and this is a difficult experimental task.

These considerations appear to favor method (a) in terms of reliability. Therefore the present proposal is based on method (a) with the additional benefit provided by a very-low-loss wall coating and by the bellows system used, which allows a large dynamical range (Δ_2/Δ_1) while the surface area is kept constant.

3. **Features of the 'accordion system'**

The new type of 'low temperature Fomblin' (LTF) used for wall coating is a fluoropolymer of composition $CF_3O(CF_2O)_n(CF_2CF_2O)_m(OCF_2CF_2O)_kCF_3$ with n = 30.3, m = 1.5, k = 0.2 [13]. It consists only of the low-absorbing elements C, F, and O, like ordinary Fomblin, but has an 80 K lower solidification temperature of ~150 K. Thus it can be used in liquid form at low temperatures where losses due to inelastic scattering [12] and quasi-elastic scattering [14,11] are strongly reduced. In direct UCN storage measurements using method (b) we obtained $\eta \sim 5\times10^{-6}$ in the temperature range 105-150 K. This is the lowest wall loss coefficient reported so far but subject to uncertainty. Since no reliable extrapolation to τ_n seemed possible in these experiments, the η-value was calculated from the τ_{st}^{-1} vs. γ slope assuming intersection at the world average value $\tau_n \sim 886$ s. The largest storage lifetime obtained in these experiments was 872 s, which is <2% away from 886 s [11].

The new system, shown schematically in Fig. 1, uses trap size variation as in method (a). A 56 cm OD bellows with stretched length 125 cm is fitted into a horizontal cylindrical vacuum chamber. The system allows gap-free volume changes by a stepper motor moving with precision ~0.01 mm. The wide volume range (and therefore also of λ and ν) by a factor >25 is achieved by insertion of a 'spacer' which reduces the minimum usable separation of the vertical walls to ~1 cm. A prominent feature of this device is the absence of changes of wall area, or of its distribution over height. As shown below, this is an essential condition for straight-line dependence of τ_{st}^{-1} on ν, independently of the energy dependence of reflection loss as well as of the UCN spectrum. An Al transmission foil (Fig. 1) provides a low-energy cutoff such that all admitted UCN are able to reach the roof.

The interior storage volume surface will be coated with LTF oil at the measurement temperature, which can be chosen in the liquid or solid range. The oil is condensed from vapor transported from a heated reservoir in low-pressure He gas (feature *A* in Fig. 1) and the coating is easily refreshed. Uniformity of surface temperature is achieved by embedding the ‘bellows volume’ in a secondary vacuum vessel kept at a temperature variable between ~100 and 200 K. The thermal contact can be improved by low-pressure He gas.

Fig. 1 shows the principle of highly leak-tight UCN and oil vapor shutters. Residual leakage can be checked with helium gas.

“Scaling” will be used for trap loading, storage and emptying, i.e. all time periods will be proportional to volume V, and therefore strictly $\sim \lambda$.

4. **Calculations**

Details of the analysis of the ‘accordion-system’ have been presented elsewhere [15]. The following assumptions were made.

(i) The initial spectrum of UCN entering the system is a Maxwell spectrum cut from above by the critical energy for LTF at ~150 K (= 120 cm in units of E/mg with the neutron mass m and gravitational constant g [11]). From below there is a smooth cutoff due to Al foil transmission. Referring all UCN jump heights to the trap center at $h = 0$, the energy range of stored UCN is 30 cm $< h_o < h_{cr}$ with $h_{cr} = 120$ - OD/2 = 92 cm, where OD is the outer bellows diameter.

(ii) A loss coefficient $\eta = 5\times10^{-6}$ was assumed but this value and the associated model of a potential-step wall affect only the calculated storage lifetimes but have very little influence on quantities like collision rate ν or trap loading/emptying time constants.

(iii) Transmission times and transmission losses between UCN valve and source or detector were neglected.

(iv) For given height, the UCN spectra and densities were assumed to be uniform over the lateral trap extension at all times, even during trap loading and emptying. We plan to use Monte Carlo simulations to check the validity of this assumption.

(v) In most calculations a long storage time was used to make sure that averaging of loss rates (as in Eq. (2)) and of ν or similar quantities is justified even for long storage periods. For the largest volume $V_{\max}$ (fully stretched bellows) we chose: for loading $t_f = 200$ s; for storage $t_1 = 300$ s (short storage time), $t_2 = 2300$ s (long storage time), and for emptying $t_e = 150$ s.

(vi) For smaller accordion volumes all times were reduced by the factor $\lambda_{\max}/\lambda = V_{\max}/V$.

(vii) The interior trap surface is constant.

(viii) Gravity was taken into account.

(ix) UCN loss by leakage through the closed valves was neglected. It adds to the reflection loss, and calculations confirm that it does not change the extrapolated end point τ_n.

(x) Residual gas loss was neglected although, as in [5,6,8-10], the trap cannot be pumped during storage. It will be baked in vacuum and the residual gas composition will be monitored to determine a possible correction to τ_n.

The most important result is: Due to constant trap surface and application of the "scaling technique" for different trap volumes V, the dependence of UCN storage loss rate τ_{st}^{-1} *versus* collision rate ν becomes a straight line. The same is true if we plot the mean τ_{st}^{-1} values from (2) vs. $\langle\nu\rangle$, the mean values for t_1 and t_2. Here the mean values are taken for the interval from $t_1 = 300$ s to $t_2 = 2300$ s (and scaled down for $V < V_{\max}$). The high degree of linearity is shown in Fig. 2 where linear fits for different subgroups of points for volumes in the range $V_{\max}/V = 1$ to 27 give τ_n extrapolations within 0.5 s of the initial input value 886 s. Even the four low-volume points farthest from the *y*-intersection (window on right) show no deviation exceeding the anticipated accuracy of the proposed τ_n experiment. Using a wall loss model with a velocity dependence of losses very different from the step function model, and adding gaps, we obtained the same degree of linearity.

In contrast to a plot vs. γ (as for method (b)), calculation of collision rates ν does not require a specific reflection loss model. However, it does require knowledge of the UCN spectrum and its change during a cycle, which is a difficult experimental task. To approximate ν by a quantity based entirely on measured quantities we use the definition of inverse mean free path $\lambda^{-1} = N\nu/\Phi = S/4V$ where the integral flux is $\Phi = N\langle v\rangle$ and the velocity is averaged over volume and spectrum. Since the trap surface area S is constant, $\lambda^{-1} \sim V^{-1}$ and the collision rate can be expressed as

$$\nu \sim \Phi/NV \sim \gamma_e. \qquad (3)$$

The first proportionality is exact. The second form is a close approximation in terms of loss-corrected efflux rate $\gamma_e = \gamma_{et} - \tau_n^{-1} - \tau_w^{-1} = \gamma_{et} - \tau_{st}^{-1}$, where γ_{et} is the initial efflux rate. γ_e can be represented by directly measurable quantities: total counts N; measured storage lifetimes; and count-rates (per s) right after opening the UCN valve at the end of storage times t_1 and t_2. A plot of τ_{st}^{-1} vs. γ_e/γ_{e0} shows the same degree of linearity as the plot vs. ν/ν_0 in Fig. 2. However, in practice the measurement of time-dependent efflux count-rates may be complicated by the guide section between valve and detector, which gives rise to time delay and losses.

5. **Conclusions**

A new ‘accordion-type’ UCN storage system with low-loss ‘low temperature Fomblin‘ coating is proposed. Analysis suggests its suitability for a neutron lifetime experiment with precision < 1s. This is mainly due to the fact that the trap surface area and its distribution over height remain constant while the volume is changeable in a wide range. Combination with the “scaling technique” of Ref. [6] ensures that the extrapolation from measured storage lifetimes to the lifetime for β-decay is almost exactly linear and therefore reliable. This is true under gravity and for any energy dependence of wall losses and any spectrum of stored UCN.

We are grateful to L. Bondarenko, M. Daum, A. Kharitonov and V. Shvetsov for invaluable contributions to the LTF-project and acknowledge the support by the U.S. Department of Energy grant DE-FG02-02ER45970.

References

1 I.S. Towner, J.C. Hardy, in *Physics beyond the Standard Model* (World Scientific 1999).

2 A. Bertin, M. Capponi *et al.*, Phys. Lett. B 199 (1987) 108.

3 R.E. Lopez, M.S. Turner, Phys. Rev. D **59** (1999) 103502-1.

4 B.G. Yerozolimsky, Nucl. Instr, Meth. Phys. Res. A **440** (2000) 491.

5 V.I. Morozov, Nucl. Instr. Meth. Phys. Res. A**284** (1989) 108.

6 W. Mampe *et al.*, Phys. Rev. Lett. **63** (1989) 593.

7 V. Nesvizhevsky *et al.*, Sov. Phys. JETP **75** (1992) 405.

8 W. Mampe *et al.*, Sov. Phys. JETP **57** (1993) 82.

9 S. Arzumanov *et al.*, in ISINN-5, *Neutron Spectroscopy, Nuclear Structure, Related Topics*, Dubna, E3-97-213, 1997, p. 53.

10 A. Pichlmaier *et al.*, Nucl. Instr, Meth. Phys. Res. A **440** (2000) 517.

11 P. Geltenbort *et al.*, *Preparation of neutron lifetime experiment using 'low temperature Fomblin'*, ISINN-11, 11-th International Seminar on Interaction of Neutrons with Nuclei, Dubna, 2003, http://surface.phys.uri.edu/Publications-Steyerl/n-lifetime.2003.pdf .

12 Yu.N. Pokotilovski, Nucl. Instr. Meth. Phys. Res. A **425** (1999) 320; JETP **96** (2003) 172.

13 Trademark "POM" supplied by the Perm' branch of the Russian Scientific Center of Applied Chemistry, Moscow.

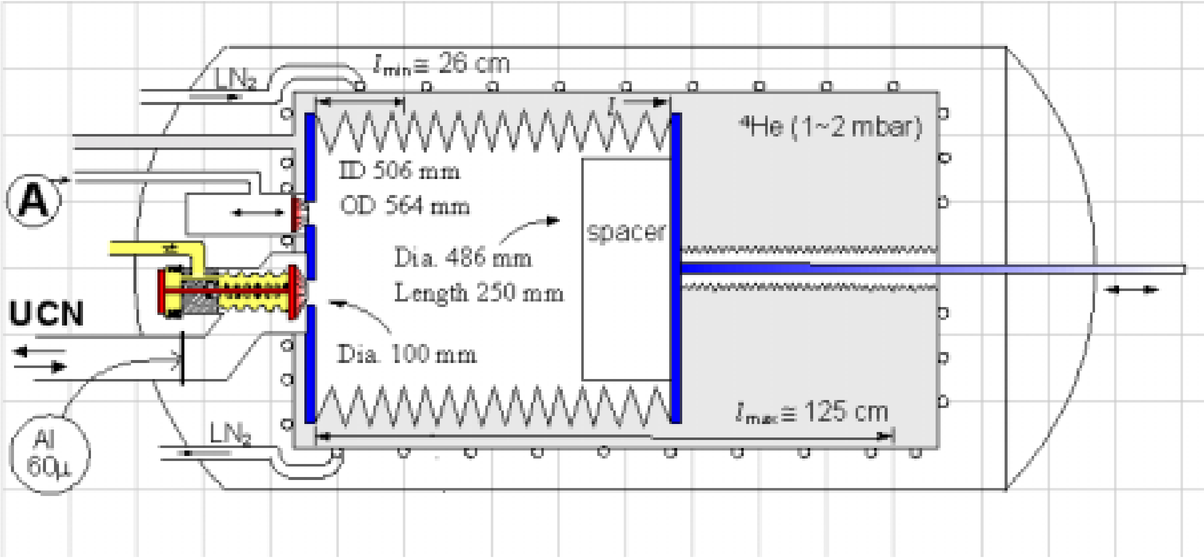
14 A. Steyerl, B. Yerozolimsky *et al*., Eur. Phys. J. B **28** (2002) 299; ISINN-10, 10-th International Seminar on Interaction of Neutrons with Nuclei, Joint Inst. of Nuclear Research, Dubna, 2002, E3-2003-10, p. 388.

15 B. Yerozolimsky *et al*., *'Low temperature Fomblin' coated accordion-like UCN storage system for neutron lifetime experiment*, ISINN-12, 12-th International Seminar on Interaction of Neutrons with Nuclei, Dubna, 2004.

Figure Captions

Fig. 1: Schematic view of the “accordion” system. It contains a UCN trap whose surface area remains constant while the volume is changeable by a factor 27. The inner surface will be coated with ‘Low Temperature Fomblin’ at temperatures in the range from 100-220 K to provide a low-loss UCN storage system for a neutron lifetime measurement.

Fig. 2: Test of linearity of a plot of inverse storage lifetime, τ_{st}^{-1}, versus relative wall collision rate ν/ν_0. For the largest trap volume V_{max}, $\nu_0 = 51.9$ Hz. The τ_{st}^{-1}-values are mean values for the storage interval from $t_1 = 300$ s to $t_2 = 2300$ s (for the largest volume). The collision rates are mean values for the same time interval. The windows show results of linear fits for subgroups of points (counting from the left): bottom: 1-4; left: 1-5; top: 1-10; right: 7-10. Extrapolated y-axis intersections agree within 0.3 s with the value $\tau_n = 886$ s used in all calculations. The error ranges reflect residuals and do not include counting statistics.



l_min ≅ 26 cm
LN2
l
4He (1~2 mbar)
A
ID 506 mm
OD 564 mm
spacer
Dia. 486 mm
Length 250 mm
UCN
Dia. 100 mm
l_max ≅ 125 cm
LN2
Al
60μ

Fig. 1

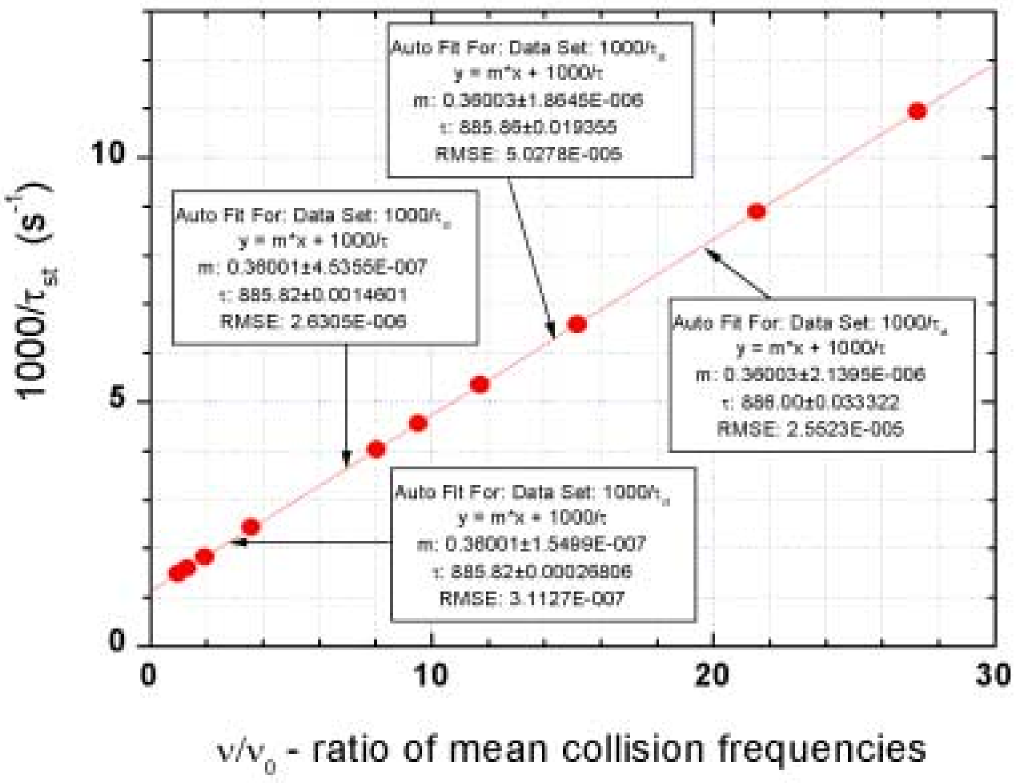

Auto Fit For: Data Set: 1000/τ_s
y = m*x + 1000/τ
m: 0.36003±1.8645E-006
τ: 885.86±0.019355
RMSE: 5.0278E-006
Auto Fit For: Data Set: 1000/τ_c
y = m*x + 1000/τ
m: 0.36001±4.5355E-007
τ: 885.82±0.0014601
RMSE: 2.6305E-006
Auto Fit For: Data Set: 1000/τ_a
y = m*x + 1000/τ
m: 0.36003±2.1395E-006
τ: 886.00±0.033322
RMSE: 2.5623E-005
Auto Fit For: Data Set: 1000/τ_d
y = m*x + 1000/τ
m: 0.36001±1.5489E-007
τ: 885.82±0.00026806
RMSE: 3.1127E-007
1000/τ_st (s^-1)
0
5
10
0
10
20
30
ν/ν_0 - ratio of mean collision frequencies

Fig. 2

DOE F 1332.16
(10-90)
All other editions
are obsolete

OMB Control No.
1910-1400
Burden Disclosure
Statement on back

U.S. DEPARTMENT OF ENERGY

UNIVERSITY CONTRACTOR, GRANTEE, AND COOPERATIVE AGREEMENT RECOMMENDATIONS FOR ANNOUNCEMENT AND DISTRIBUTION OF DOCUMENTS

(See Instructions on Reverse Side)

1. DOE Report No.	2. DOE Contract No.	3. DOE Funding Office	4. OSTI UC Category No.
DOE/ER/45970-12	DE-FG02-02ER45970	Materials Sciences	

5. Title ‘Low Temperature Fomblin’ coated accordion-like UCN storage system for neutron lifetime experiment, by B. Yerozolimsky, A. Steyerl, O. Kwon, V. Luschikov, A. Strelkov, P. Geltenbort, N. Achiwa, A. Pichlmaier, P. Fierlinger

6. Type of Document (“x” one). Two completely legible reproducible copies should be transmitted.

a. Scientific and technical report: ☐ monthly ☐ quarterly ☐ annual ☐ final ☐ topical ☐ other

b. Conference paper: Name of conference (no abbreviations) ISINN-12, International Seminar on Interaction of Neutrons with Nuclei

Location (city/st/ctry) Dubna, Russia

Date (mo/day/yr) 5/26-29/2004 Sponsor Joint Institute for Nuclear Research, Dubna

Contents: ☐ proceedings ☐ viewgraphs ☒ paper ☐ poster sessions

for proceedings

c. Computer Media:

Document is accompanied by ☐ magnetic tape(s)

☐ diskette(s)

☐ other (specify)

d. Other (e.g., journal article, manuscript, manual, etc.) (specify)

7. Patent Information

a. Does this information product disclose any new equipment, process, or material? ☒ No ☐ Yes If so, identify page nos.

b. Has an invention disclosure been submitted to DOE covering any aspect of this information product? ☒ No ☐ Yes
If so, identify the DOE (or other) disclosure number and to whom the disclosure was submitted.

c. Are there any patent-related objections to the release of this information product? ☒ No ☐ Yes If so, state these objections.

d. Does this material contain Proprietary information? ☒ No ☐ Yes

8. Additional information, remarks, and special handling instructions. (Continue on separate sheet, if necessary.)

9. Submitted by (Name and Position) (Please print or type)		Phone
Albert Steyerl, Prof. of Physics		401-874-2204
Organization	**Signature**	**Date**
University of Rhode Island	A. Steyerl	8/10/2004

‘LOW TEMPERATURE FOMBLIN’ COATED ACCORDION-LIKE UCN STORAGE SYSTEM FOR NEUTRON LIFETIME EXPERIMENT

B. Yerozolimsky[1], A. Steyerl[2], O. Kwon[2], V. Luschikov[3], A. Strelkov[3], P. Geltenbort[4], N. Achiwa[5], A. Pichlmaier[6], P. Fierlinger[6]

[1] Harvard University, Cambridge, MA, USA; [2] University of Rhode Island, Kingston, RI, USA; [3] Joint Institute for Nuclear Research, Dubna, Russia; [4] Institut Laue Langevin, Grenoble, France; [5] Osaka University, Japan; [6] Paul Scherrer Institut, Villigen, Switzerland

Key words: neutron lifetime; ultracold neutrons

A new type of per-fluorinated polymer, ‘Low Temperature Fomblin’, has been tested as a wall coating in an ultracold neutron (UCN) storage experiment using a gravitational storage system. The data show a UCN reflection loss coefficient η as low as ~ 5×10^{-6} in the temperature range 105 - 150 K. We plan to use this oil in a new type of neutron lifetime measurement, where a bellows system (‘accordion’) enables to vary the trap size in a wide range while the total surface area and distribution of surface area over height remain constant. These unique characteristics, in combination with application of the scaling technique developed by W. Mampe et al. in 1989, ensure exact linearity for the extrapolation from inverse storage lifetimes to the inverse neutron lifetime. Linearity holds for any energy dependence of loss coefficient $\mu(E)$. Using the UCN source at the Institut Laue Langevin we expect to achieve a lifetime precision below ±1 s.

1. **Introduction**

Particle decay data indicate that the Cabibbo-Kobayashi-Maskawa matrix may deviate from unitarity (presently at the 2.7-sigma level [1]). This question depends critically on the up-down quark mixing amplitude V_{ud}, which is determined most sensitively by the neutron lifetime τ_n and the neutron decay asymmetry coefficient A. A reliable, precise value of τ_n will also help to refine models of astrophysics [2] and cosmology [3]. The current world average is $\tau_n = 885.7\pm0.8$s [4]. We propose a new τ_n measurement with a precision below 1s, using UCN storage. Wall losses are minimized by the use of ‘low-temperature Fomblin’, and the notoriously difficult extrapolation from storage lifetimes to τ_n is made more reliable by the novel use of an accordion-like storage vessel. In this system, the surface area and its distribution over height remain constant while the volume is changeable in a wide range. Combination with the scaling technique of Mampe *et al.* [6] ensures that the extrapolation function becomes strictly linear for any shape of UCN spectrum in the trap and for any energy dependence of reflection loss coefficient μ. Moreover, no correction for gravity is required. These unique features distinguish this method from all previous τ_n experiments based on UCN storage in material traps [5-10].

2. **Basic considerations**

In UCN-storage based τ_n experiments it is crucial that any non-decay loss due to wall collisions, gaps and the residual gas are reliably subtracted from the total loss rate. In the elementary theory of wall reflection loss the interaction with the wall atoms is described by a step-function barrier determined by the optical (or mean Fermi) potential U–iW. For an isotropic UCN distribution the mean loss probability per bounce is given by $\mu(E) = 2\eta\{(U/E)\arcsin(E/U)^{1/2} - [(U-E)/E]^{1/2}\}$, where $\eta = W/U$ and E is the neutron kinetic energy at the impact point. The wall loss probability per second is

$$\tau_w^{-1} = \langle \nu\mu(E) \rangle \qquad (1)$$

where ν is the wall collision rate for a UCN and the average is taken over the UCN spectrum and trap surface. Neglecting gravity, $\nu = \mathrm{v}/\lambda$ can be expressed by the mean UCN velocity v and the mean free path $\lambda = 4V/S$. λ is independent of v and determined only by the total trap surface S and volume V. As shown in [6] this gas-kinetic result is also valid under gravity provided the trap geometry has a horizontal plane of symmetry (as in the 'accordion-trap' discussed below) and all UCN have enough energy to reach the highest point(s) in the trap. For gravitationally bent paths we use the straightforward definition $1/\lambda$ = total wall collision rate $N\nu$ divided by the volume-integrated UCN flux Φ in the trap (see Sec. 4).

However, gravity does induce an important difference. For our trap geometry, the total UCN number N is not exactly proportional to volume V, even for identical spectra. As a consequence we will plot storage data vs. ν, not $1/\lambda$, to obtain a linear dependence.

If we are sure that a single constant (η) can be factored out of the function $\mu(E)$, Eq. (1) may also be written in the form $\tau_w^{-1} = \eta\gamma$, where γ is defined as $\gamma = \langle(\mathrm{v}/\lambda)(\mu/\eta)\rangle$, averaged over spectrum and surface. However, this is strictly justified only for a smooth, uniform wall without surface contamination and/or microstructure (cracks, roughness etc.), which can be represented by a potential step function.

In an experiment we measure the numbers $N(t_1)$, $N(t_2)$ of UCN counted after storage times t_1 and t_2. Although for a broad UCN spectrum the decay curve is non-exponential one can define a mean storage lifetime for time interval t_1, t_2 in the form

$$\tau_{st} = (t_2 - t_1)/\ln[N(t_1)/N(t_2)] \qquad (2)$$

with $\tau_{st}^{-1} = \tau_n^{-1} + \tau_w^{-1}$ + other losses.

Three different methods have been used to extract τ_n from storage experiments.

(a) In Refs. [5,6,10], the vessel geometry was changed to vary λ and extrapolate the measured dependence $\tau_{st}^{-1}(\lambda^{-1})$ to $\lambda^{-1} \to 0$.

(b) In Ref. [7], the storage lifetime was measured for different intervals of UCN energy, corresponding to different mean γ-values, and the dependence $\tau_{st}^{-1}(\gamma)$ was extrapolated to $\gamma \to 0$.

(c) In Ref. [9], storage lifetimes were measured together with the flux of UCN thermally up-scattered at the trap walls. This provided a further handle on the separation of wall losses from beta-decay.

Restricting the discussion to the extrapolation methods (a) and (b), the accuracy and reliability of the y-axis intersection is determined by several criteria:

i) First of all, the reliability of the extrapolation law $\tau_{st}^{-1}(\lambda^{-1})$ or $\tau_{st}^{-1}(\gamma)$ (or e.g. $\tau_{st}^{-1}(\nu)$) used to bridge the gap Δ_1 from $(\lambda^{-1})_{min}$ or γ_{min} (or similar x-variables) to zero.

ii) The maximum x-range Δ_2 between $(\lambda^{-1})_{min}$ and $(\lambda^{-1})_{max}$ (or similar) accessible to the experiment. If the dependence is linear and the statistical uncertainties are constant, the precision of the fitted y-intersection (τ_n^{-1}) is determined by the ratio Δ_2/Δ_1, and independent of the size of the gap Δ_1 (the distance of τ_{st} from τ_n).

iii) In addition to the experimental y-errors the uncertainties of calculated mean x-values like γ must be taken into account [11]. They include spectral and model uncertainties, most critically those related to the assumption of a step-function potential for the wall.

Method (a) ensures a linear dependence *if* all τ_{st} data are obtained with the same UCN energy spectrum at equivalent times of a storage cycle, so that only λ changes when the trap geometry is changed. The linearity can be checked for the experimental data, and the reliability of this test will also improve with increasing Δ_2/Δ_1. To ensure identical spectra in storage measurements with different λ, Pendlebury, Mampe *et al.* developed a scaling procedure where all time intervals Δt are chosen proportional to λ [6]. In this case, the total number of wall collisions in equivalent time intervals is the same, and therefore the spectra and spectral changes are practically the same for cycles with different λ-values. In Ref. [6], only small corrections were needed to take into account the 'loading effect' (essentially the role of β-decay during trap loading and emptying) and gravity. Both corrections are essentially reduced for the system described below.

Method (b) relies on the validity of the assumed energy dependence of the wall loss. The calculation of γ-values is directly based on the step function potential model for the wall. Experiments show that for low-absorbing materials like per-fluorinated polymers at low temperature or solid oxygen the loss coefficient is significantly higher than calculated for a clean surface [12]. Moreover, in a ~10^{-5} mbar vacuum at ~120 K a clear deterioration of τ_{st} by several s was observed over time periods of hours to days [11], possibly due to surface contamination from the residual gas. Besides, calculation of the γ-values required for the energy extrapolation method depends on the UCN spectra. Therefore all spectral changes over a cycle must be known, including Doppler shifts due to trap rotation, and this is a difficult experimental task.

These considerations appear to favor method (a) in terms of reliability. Therefore the present proposal is based on method (a) with the additional benefit provided by a very-low-loss wall coating and by the bellows system used, which allows a large dynamical range (Δ_2/Δ_1) while the surface area is kept constant.

3. Features of the 'accordion system'

The new type of 'low temperature Fomblin' (LTF) used for wall coating is a fluoropolymer of composition $CF_3O(CF_2O)_n(CF_2CF_2O)_m(OCF_2CF_2O)_kCF_3$ with n = 30.3, m = 1.5, k = 0.2 [13]. It consists only of the low-absorbing elements C, F, and O, like ordinary Fomblin, but has an 80 K lower solidification temperature of ~150 K. Thus it can be used in liquid form at low temperatures where losses due to inelastic scattering [12] and quasi-elastic scattering [14,11] are strongly reduced. In direct UCN storage measurements using method (b) we obtained $\eta \sim 5\times10^{-6}$ in the temperature range 105-150 K. This is the lowest wall loss coefficient reported so far but subject to uncertainty. Since no reliable extrapolation to τ_n seemed possible in these experiments, the η-value was calculated from the τ_{st}^{-1} vs. γ slope assuming intersection at the world average value $\tau_n \sim 886$ s. The largest storage lifetime obtained in these experiments was 872 s, which is <2% away from 886 s [11].

The new system, shown schematically in Fig. 1, uses trap size variation as in method (a). A 56 cm OD bellows with stretched length 125 cm is fitted into a horizontal cylindrical vacuum chamber. The system allows gap-free volume changes by a stepper motor moving with precision ~0.01 mm. The wide volume range (and therefore also of λ and ν) by a factor >25 is achieved by insertion of a 'spacer' which reduces the minimum usable separation of the vertical walls to ~1 cm. A prominent feature of this device is the absence of changes of wall area, or of its distribution over height. As shown below, this is an essential condition for straight-line dependence of τ_{st}^{-1} on ν, independently of the energy dependence of reflection loss as well as of the UCN spectrum. An Al transmission

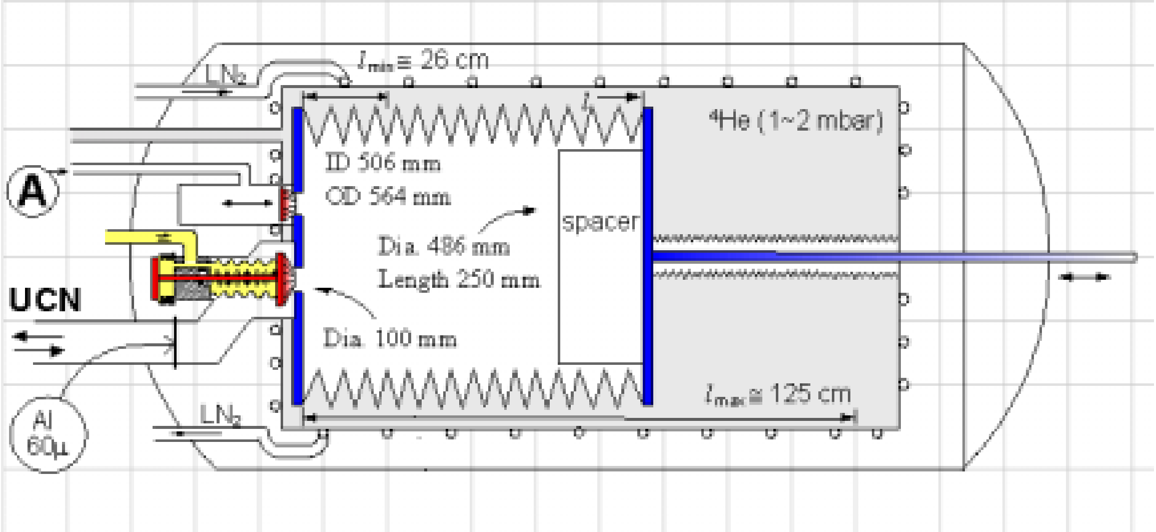


Fig. 1: Schematic view of the "accordion" system. It contains a UCN trap whose surface area remains constant while the volume is changeable by a factor 27. The inner surface will be coated with 'Low Temperature Fomblin' at temperatures in the range from 100-220 K to provide a low-loss UCN storage system for a neutron lifetime measurement.

foil (Fig. 1) provides a low-energy cutoff such that all admitted UCN are able to reach the roof.

The interior storage volume surface will be coated with LTF oil at the measurement temperature, which can be chosen in the liquid or solid range. The oil is condensed from vapor transported from a heated reservoir in low-pressure He gas (feature *A* in Fig. 1) and the coating is easily refreshed. Uniformity of surface temperature is achieved by embedding the 'bellows volume' in a secondary vacuum vessel kept at a temperature variable between ~100 and 200 K. The thermal contact can be improved by low-pressure He gas.

Fig. 1 shows the principle of highly leak-tight UCN and oil vapor shutters. Residual leakage can be checked with helium gas.

"Scaling" will be used for trap loading, storage and emptying, i.e. all time periods will be proportional to volume *V*, and therefore strictly ~ λ.

4. **Calculations**

To analyze the 'accordion-system' we made the following assumptions:

(i) The initial spectrum of UCN entering the system is a Maxwell spectrum cut from above by the critical energy for LTF at ~150 K (= 120 cm in units of E/mg with the neutron mass m and gravitational constant g [11]). From below there is a smooth cutoff due to Al foil transmission. Referring all UCN jump heights to the trap center at $h = 0$, the energy range of stored UCN is 30 cm $< h_o < h_{cr}$ with $h_{cr} = 120 - \mathrm{OD}/2 = 92$ cm, where OD is the outer bellows diameter.

(ii) A loss coefficient $\eta = 5\times10^{-6}$ was assumed but this value and the underlying model of a potential-step wall affect only the calculated storage lifetimes but have very little influence on quantities like collision rate ν or trap loading/emptying time constants.

(iii) Transmission times and transmission losses between UCN valve and source or detector were neglected.

(iv) For given height, the UCN spectra and densities were assumed to be uniform over the lateral trap extension at all times, even during trap loading and emptying. We plan to use Monte Carlo simulations to check the validity of this assumption.

(v) In most calculations a long storage time was used to make sure that averaging of loss rates (as in Eq. (2)) and of ν or similar quantities is justified even for long storage periods. For the largest volume $V_{\max}$ (fully stretched bellows) we chose: for loading $t_f = 200$ s; for storage $t_1 = 300$ s (short storage time), $t_2 = 2300$ s (long storage time), and for emptying $t_e = 150$ s.

(vi) For smaller accordion volumes all times were reduced by the factor $\lambda_{\max}/\lambda = V_{\max}/V$.

(vii) The interior trap surface is constant.

(viii) Gravity was taken into account.

(ix) UCN loss by leakage through the closed valves was neglected. It adds to the reflection loss, and calculations confirm that it does not change the extrapolation to τ_n.

(x) Residual gas loss was neglected although, as in [5,6,8-10], the trap cannot be pumped during storage. It will be baked in vacuum and the residual gas composition will be monitored to determine a possible correction to τ_n.

We need the following quantities to calculate λ^{-1}, ν and the wall loss [15]:

(i) The total number of UCN that can be counted at cycle end after having escaped losses during storage (for time t), loading, and emptying. The loss is due to β-decay (at a rate $\gamma_\beta = \tau_n^{-1}$) and reflection loss with rate $\gamma_w = \tau_w^{-1}$, averaged over the trap surface and over the spectrum with phase-space density $\rho(h_o)$ [$\mathrm{m}^{-3}(\mathrm{m/s})^{-3}$] for UCN with energy h_o [16]:

$$N = 2\pi(2g)^{3/2} \int dh_o \rho(h_o)\, f_{fe}(h_o) \exp[-(\gamma_\beta+\gamma_w)t]\, f_N(h_o) \tag{3}$$

with the spectral depletion factor for loading plus emptying

$$f_{fe}(h_o) = [1-\exp(-\gamma_{et}t_f)](\gamma_e/\gamma_{et})^2[1-\exp(-\gamma_{et}t_e)], \tag{4}$$

the density integral over the trap volume V

$$f_N(h_o) = \int dV\, (h_o-h)^{1/2} \tag{5}$$

and the loading/emptying rates γ_e (excluding losses) and $\gamma_{et} = \gamma_e+\gamma_\beta+\gamma_w$ (including losses). The 0.1% difference of wall loss rate with open vs. closed UCN valve can be neglected.

(ii) The volume-integrated UCN flux $\Phi = N\langle v\rangle$ is given in the form of Eq. (3) with $f_N(h_o)$ replaced by

$$f_\Phi(h_o) = (2g)^{1/2} \int dV\, (h_o-h). \tag{6}$$

Due to the trap symmetry about $h=0$, $f_\Phi(h_o) = (2g)^{1/2}h_oV$, thus

$$\Phi = 8\pi g^2 V \int dh_o\, \rho(h_o)\, h_o\, f_{fe}(h_o) \exp[-(\gamma_\beta+\gamma_w)t]. \tag{7}$$

(iii) The total wall collision rate $N\nu$ is given in the form (3) with $f_N(h_o)$ replaced by the surface integral

$$f_\nu(h_o) = \tfrac{1}{4}(2g)^{1/2} \int dS\, (h_o-h) = \tfrac{1}{4}(2g)^{1/2}\, h_oS, \tag{8}$$

thus

$$N\nu = 2\pi g^2 S \int dh_o\, \rho(h_o)\, h_o\, f_{fe}(h_o) \exp[-(\gamma_\beta+\gamma_w)t]. \tag{9}$$

(iv) The total wall reflection loss Λ is also given in the form (3) with $f_N(h_o)$ replaced by the surface integral with any energy dependence of loss coefficient $\mu(h_o-h)$,

$$f_\Lambda(h_o) = \tfrac{1}{4}(2g)^{1/2} \int dS\, (h_o-h)\, \mu(h_o-h), \tag{10}$$

thus

$$\Lambda = 2\pi(2g)^{3/2}\int dh_o\rho(h_o)\, f_{fe}(h_o) \exp[-(\gamma_\beta+\gamma_w)t]\, f_\Lambda(h_o). \tag{11}$$

(v) The total trap loading/emptying rate $N\gamma_e$ is given in the form (3) with $f_N(h_o)$ replaced by the surface integral over the circular UCN entrance/exit cross section S_{ef},

$$f_\gamma(h_o) = \tfrac{1}{4}(2g)^{1/2} \int dS_{ef}\,(h_o-h) = \tfrac{1}{4}(2g)^{1/2}\,(h_o-h_{ef})\,S_{ef}, \tag{12}$$

where $h_{ef} = -18.2$ cm is the aperture center height; thus

$$\gamma_e = 2\pi g^2\, S_{ef} \int dh_o\, \rho(h_o)\,(h_o-h_{ef})\, f_{fe}(h_o)\, \exp[-(\gamma_\beta+\gamma_w)t]\, /\, N. \tag{13}$$

Using the definition

$$\lambda^{-1} = N\nu/\Phi, \tag{14}$$

equations (7) and (9) confirm that the gas-kinetic result $\lambda^{-1} = S/4V$ holds also under gravity [6], provided the trap is symmetrical and all UCN have enough energy to reach the roof. Since S = constant, $\lambda^{-1} \sim V^{-1}$. Furthermore, as a result of the "scaling technique" for different trap volumes V, all surface integrals, and thus also $N\nu$, Λ, and $N\gamma_e$ become independent of V, apart from β-decay which depends on the cycle length. However, this dependence cancels out in a plot of total UCN storage loss rate $\tau_{st}^{-1} = \gamma_\beta + \Lambda/N$ *versus* ν.

The same is true if we plot the mean τ_{st}^{-1} values from (2) vs. $<\nu>$, the mean values for t_1 and t_2. Here the mean values are taken for the interval from $t_1 = 300$ s to $t_2 = 2300$ s (and scaled down for $V < V_{max}$). The high degree of linearity is shown in Fig. 2 where linear fits for different subgroups of points for volumes in the range $V_{max}/V = 1$ to 27 give τ_n extrapolations within 0.5 s of the initial input value 886 s. Even the four low-volume points farthest from the *y*-intersection (window on right) show no deviation exceeding the anticipated accuracy of the proposed τ_n experiment. Using a wall loss model with a velocity dependence of losses very different from the step function model, and adding gaps, we obtained the same degree of linearity.

In contrast to a plot vs. γ (as for method (b)), calculation of ν values does not require a specific reflection loss model. However, it does require knowledge of the UCN spectrum and its change during a cycle, which is a difficult experimental task. To approximate ν by a quantity based entirely on measured quantities we write Eq. (14) in the form $\nu = \lambda^{-1}\Phi/N$ or, since $\lambda^{-1} \sim V^{-1}$,

$$\nu \sim \Phi/NV \sim \gamma_e. \tag{15}$$

The first proportionality is exact. The second form is a close approximation in terms of initial loss-corrected efflux rate $\gamma_e = \gamma_{et}-\gamma_\beta-\gamma_w = \gamma_{et}-\tau_{st}^{-1}$. It can be represented by a combination of directly measurable quantities: the total counts N, the measured storage lifetimes, and the count-rates (per s) right after opening the UCN valve after storage times t_1 and t_2. A plot of τ_{st}^{-1} vs. γ_e/γ_{e0} shows the same degree of linearity as the plot vs. ν/ν_0 in Fig. 2. However, in practice the measurement of time-dependent efflux count-rates may be complicated by the guide section between valve and detector, which gives rise to time delay and losses.

5. **Conclusions**

A new 'accordion-type' UCN storage system with low-loss 'low temperature Fomblin' coating is proposed. Analysis suggests its suitability for a neutron lifetime experiment with precision < 1s. This is mainly due to the fact that the trap surface area and its distribution over height remain constant while the volume is changeable in a wide range. This feature, in combination with applicability of the "scaling technique" of Ref. [6], ensures that the extrapolation from measured storage lifetimes to the lifetime for β-decay is almost exactly linear and therefore reliable. This is true under gravity and for any energy dependence of wall losses and any spectrum of stored UCN.

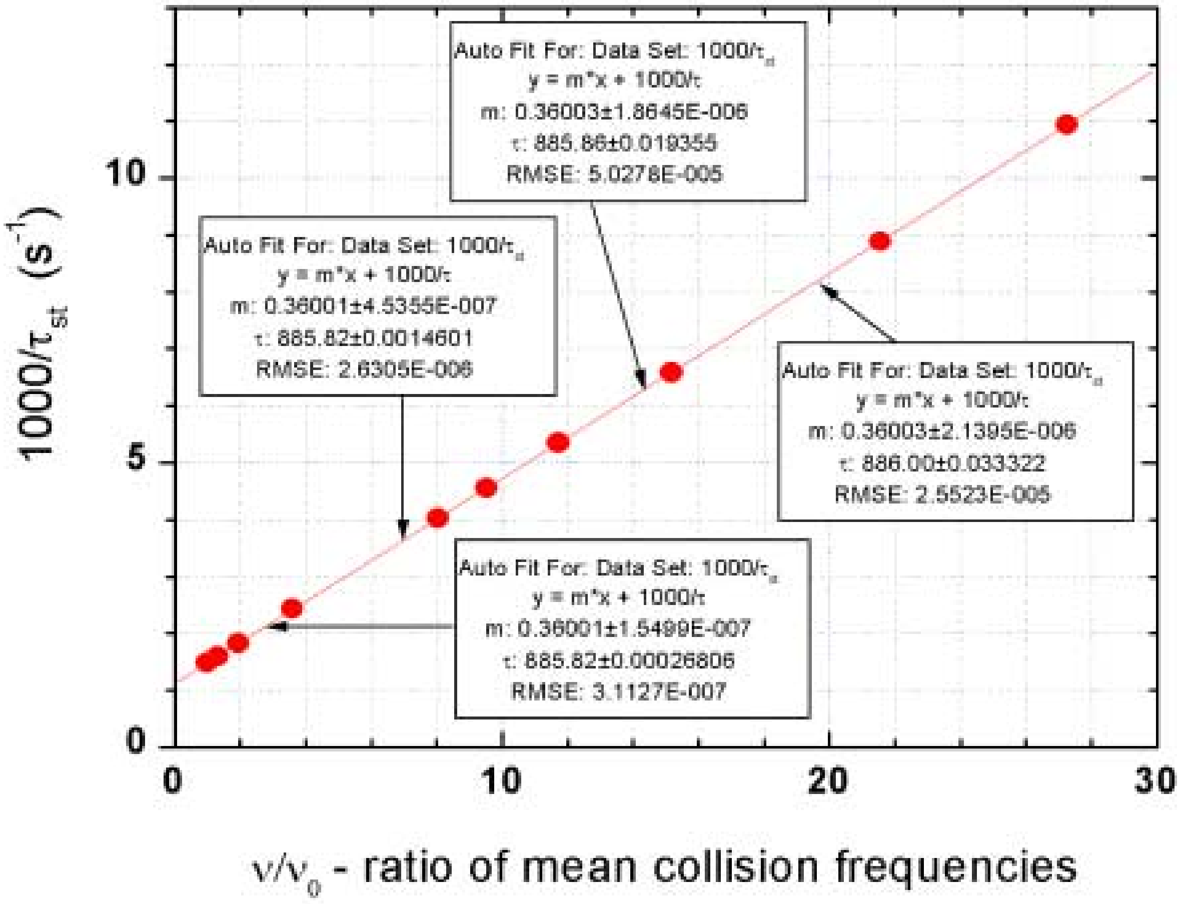


Fig. 2: Test of linearity of a plot of inverse storage lifetime, τ_{st}^{-1}, versus relative wall collision rate ν/ν_0. For the largest trap volume V_{max}, $\nu_0 = 51.9$ Hz. The τ_{st}^{-1}-values are mean values for the storage interval from $t_1 = 300$ s to $t_2 = 2300$ s (for the largest volume). The collision rates are mean values for the same time interval, calculated using (9). The windows show results of linear fits for subgroups of points (counting from the left): bottom: 1-4; left: 1-5; top: 1-10; right: 7-10. The extrapolated y-axis intersections agree within 0.3 s with the value $\tau_n = 886$ s used in all calculations. The error ranges reflect residuals and do not include counting statistics.

We are grateful to L. Bondarenko, M. Daum, A. Kharitonov and V. Shvetsov for invaluable contributions to the LTF-project and acknowledge the support by the U.S. Department of Energy grant DE-FG02-02ER45970.

1 I.S. Towner, J.C. Hardy, in *Physics beyond the Standard Model* (World Scientific 1999).
2 A. Bertin, M. Capponi *et al.*, Phys. Lett. B 199 (1987) 108.
3 R.E. Lopez, M.S. Turner, Phys. Rev. D **59** (1999) 103502-1.
4 B.G. Yerozolimsky, Nucl. Instr, Meth. Phys. Res. A **440** (2000) 491.
5 V.I. Morozov, Nucl. Instr. Meth. Phys. Res. A**284** (1989) 108.
6 W. Mampe *et al.*, Phys. Rev. Lett. **63** (1989) 593.
7 V. Nesvizhevsky *et al.*, Sov. Phys. JETP **75** (1992) 405.
8 W. Mampe *et al.*, Sov. Phys. JETP **57** (1993) 82.
9 S. Arzumanov *et al.*, in ISINN-5, *Neutron Spectroscopy, Nuclear Structure, Related Topics*, Dubna, E3-97-213, 1997, p. 53.
10 A. Pichlmaier *et al.*, Nucl. Instr, Meth. Phys. Res. A **440** (2000) 517.
11 P. Geltenbort *et al.*, *Preparation of neutron lifetime experiment using 'low temperature Fomblin'*, ISINN-11, International Seminar on Interaction of Neutrons with Nuclei, Dubna, 2003, http://surface.phys.uri.edu/Publications-Steyerl/n-lifetime.2003.pdf .

12 Yu.N. Pokotilovski, Nucl. Instr. Meth. Phys. Res. A **425** (1999) 320; JETP **96** (2003) 172.

13 Trademark “POM” supplied by the Perm’ branch of the Russian Scientific Center of Applied Chemistry, Moscow.

14 A. Steyerl, B. Yerozolimsky *et al.*, Eur. Phys. J. B **28** (2002) 299; ISINN-10, 10-th International Seminar on Interaction of Neutrons with Nuclei, Joint Inst. of Nuclear Research, Dubna, 2002, E3-2003-10, p. 388.

15 R. Golub, D. Richardson and S.K. Lamoreaux, *Ultra-Cold Neutrons*, Hilger, 1991.

16 For a Maxwell spectrum at low energy, $\rho(h_o)$ = constant, and for given energy h_o, $\rho(h_o,h)$ = constant for $h<h_o$ and =0 for $h\geq h_o$.