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# Spectral Evolution During Ultracold Neutron Storage

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**Abstract.** — In a precision measurement of the spectrum of ultracold neutrons as it evolves during storage in a “neutron bottle” we have observed an indication of a surprising heating by  $\approx 10^{-10}$  eV, occurring during the initial several 100 s of storage. We have not found any simple explanation. The data are, however, consistent with unconventional ideas proposed previously by two of us.

## 1. Introduction

Over the past years, various “anomalies” have been reported in experiments with UltraCold Neutrons (UCN). Of these, best known is a hitherto unexplained enhanced loss rate for UCN stored in low-temperature traps with very small capture and inelastic scattering cross sections of the wall material [1]. Secondly, a possible slight spectral shift for UCN in a copper trap had been indicated in [2]; and third, observation of an unexpected slight line broadening in UCN diffraction on a ruled grating, as compared to the line profile for mirror reflection, has been reported in [3].

In the present work we describe the results of an experiment performed at the High-Flux Reactor of the Institut Laue-Langevin (ILL), Grenoble, measuring the time evolution of the spectrum of UCN stored in a material trap. The experimental resolution was sufficient to detect possible energy shifts by less than  $10^{-10}$  eV (which corresponds to 1 mm of variation of maximum neutron jump height in the Earth’s gravity field). This level of precision was required to test a speculative note [4] suggesting that, indeed, a small spectral shift of up to about 3 mm may occur during the initial several thousand wall reflections in a UCN trap.

Such a shift would not be understandable on the basis of inelastic scattering in the thin layer of wall material that the UCN “see” during reflection. Thermal energy transfers are typically of order 10 meV (corresponding to room temperature, at which the walls were kept in the experiments described here), and they occur at a small rate: about 1/10000 per wall collision. Such transfers result in the certain removal of the UCN from the trap. To remain trapped

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after a transfer the energy gain has to be of order  $1/10000$  meV or less, and the probability of such a small transfer by thermal motion is totally negligible. Macroscopic vibrations of the wall were reduced to an insignificant level as shown below.

## 2. Experimental Set-Up

We used one of the UCN beams from the Cold Source/Neutron Turbine Facility [5] at the ILL, with a density of about  $30 \text{ cm}^{-3}$  for UCN velocities up to  $6.0 \text{ m s}^{-1}$ , the limit of total reflection at normal incidence for stainless steel walls (the material used here for all connecting UCN guide tubes). The measuring principle was as follows: store monochromatic UCN in a trap and measure the spectrum after different storage times.

Figure 1 shows the experimental set-up: on the left the monochromator unit (system 1) and on the right the storage and spectrometer unit (system 2). The *monochromator* consists of a stainless steel cylinder of diameter 60 cm. A "fence" of height  $17.1 \pm 0.05$  cm and diameter 40 cm divides the volume into two parts: the interior chamber I which is connected to the UCN source *via* the entrance shutter, and the exterior, annular chamber II in which monochromatic UCN were accumulated during the filling time when the entrance shutter (a rotatable disk) was open. Since the fence must be jumped by UCN to enter chamber II, it cuts the spectrum from below at  $\approx 17$  neV (measured from the chamber floor level). The upper 5 cm of fence were made of 0.1 mm thick Al foil shaped like a meander to double its total length and thus the speed of filling chamber II.

The spectrum of UCN allowed to enter chamber II was cut also from above, by use of an "absorbing roof" of adjustable height. It was made of polyethylene which reflects UCN very little since its optical potential for neutrons is small and negative. The roof was grooved, and mounted on a motor shaft rotating at 1.5–3.5 Hz in order to accelerate the speed of rejection of UCN incident on it. They are removed from the trap by thermal up-scattering or by Doppler shift to energies too high for further storage. The gravitational monochromator makes use of the definition of an energy band-width by the minimum and maximum jump heights reached by UCN when they move (almost) vertically in the gravitational field. The sharpness of the lower and upper spectral edges is improved by the choice of a long barrier ("fence") defining the lower edge, and of a long exposure time of the UCN gas to the absorber defining the upper edge.

In a first set of experiments only system 1 was used. Chamber II served as the storage cell for monochromatic UCN (with energy  $\approx 17$  neV and velocity  $1.8 \text{ m s}^{-1}$  at the trap bottom) and the exterior rim of the rotor was used for energy scanning after different storage times. In order to achieve long storage lifetimes the walls of chamber II were coated with Fomblin (a fully fluorinated polyether with very small nuclear absorption for neutrons), using oil on the floor and grease on the side walls (the latter making the walls sufficiently rough to ensure fast randomization of flight directions after filling - within about 1 second). The flap-type exit valve used had a negligible leak rate when closed. After storage in system 1 the UCN fell through a vertical distance of  $\approx 1$  m before being counted in the  $^3\text{He}$  detector, whose efficiency was improved to  $\approx 70\%$  due to the gravity acceleration.

In the second set of experiments systems 1 and 2 were connected as shown in Figure 1. System 1 now served only as the monochromator for UCN which were then stored and analyzed in system 2. The UCN were transferred to the *storage and spectrometer unit* (system 2) through a beam switch which also allowed the UCN returning after storage to be guided to the detector. The storage chamber in system 2 was a flat rectangular box of size  $67 \times 67 \times 20 \text{ cm}^3$ , coated with Fomblin grease. The maximum jump height of monochromatic UCN in the trap was  $\approx 12$  cm (and the maximum velocity was  $1.5 \text{ m s}^{-1}$ ). The trap was filled and emptied through the wide

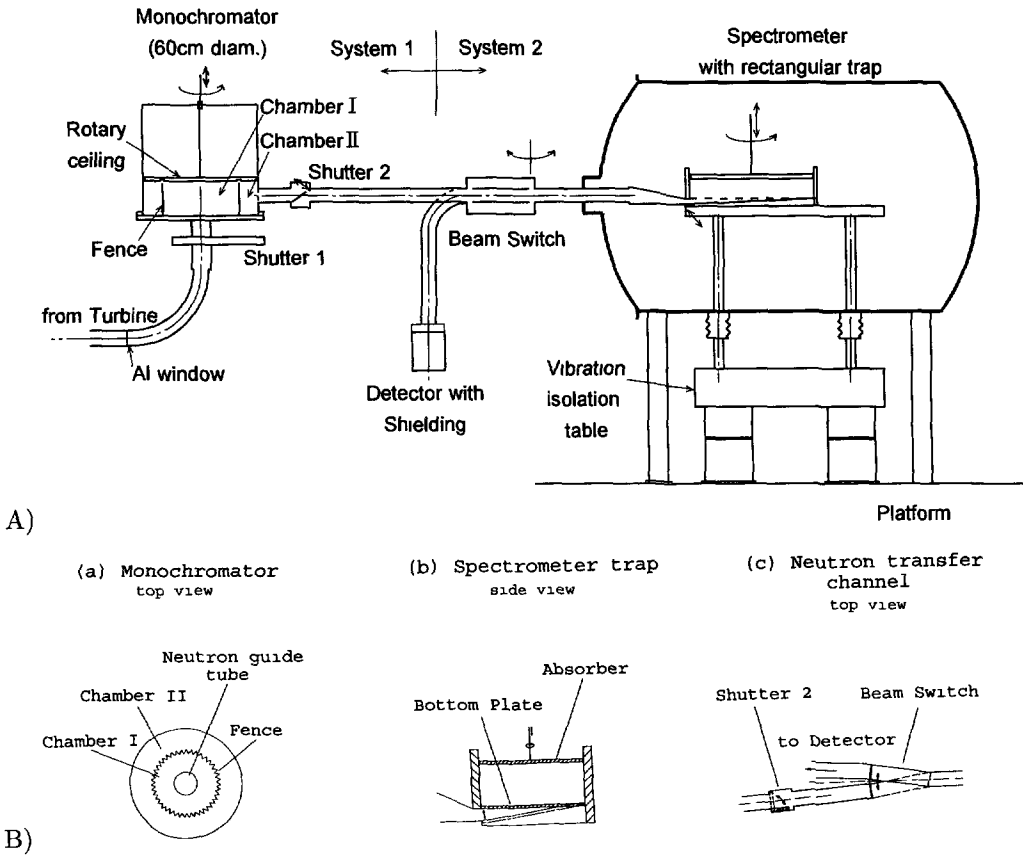


Fig 1. — A) Experimental set-up used to measure slight spectral changes for UCN stored, either in the annular region (chamber II) of the monochromator when only system 1 was used. or by the flat rectangular vessel in system 2. In either case, mono-energetic UCN were produced in the monochromator, using gravity to establish both a lower and an upper spectral cutoff. For scanning of the spectrum after various storage times the rotating absorber lid is positioned at the desired maximum jump height  $h$  for the neutron gas, left there sufficiently long for “scooping off” UCN with energies exceeding  $h$ , and then the surviving UCN are counted. Technical details are shown in the insets B): (a) the “fence” defining the lower energy cutoff in the monochromator (top view); (b) the storage chamber in system 2 (front view), and (c) the exit shutter and beam switch.

gap ( $3 \times 67 \text{ cm}^2$ ) opened by tilting the front end of the bottom plate downward. A rotatable, grooved polyethylene roof of adjustable height allowed the scanning of the UCN spectrum after storage times in the range from 100 to 800 s.

The storage chamber in system 2 rested on an air cushioned vibration isolation table and was isolated from the vacuum tank by four soft bellows as shown in Figure 1. This scheme allowed a 25 fold suppression (to  $\approx 5 \mu\text{m s}^{-1}$ ) of rms wall velocity, as compared to system 1 where fore-pump vibrations were damped only by soft supports.

Both systems were pumped to  $\leq 5 \times 10^{-5}$  mbar by turbo-molecular pumps, using (for system 1) a liquid-nitrogen cooled trap to prevent diffusion of roughing-pump oil vapour into the vacuum vessel.

### 3. Procedure of Measurements

In measurements using system 1 only, a narrow UCN spectrum was filled into chamber II for 400 s. The nominal spectral width was determined by the gap  $h_0$  ( $= 3$  mm in most runs) between the fence top and the roof position. After filling chamber II the roof was raised to  $h_s = 9$  mm where it did not interfere with the UCN gas during the storage time  $t_s$ . During the last phase of storage the roof was lowered to the desired scanning position  $h$  ( $-9 \text{ mm} \leq h \leq 9 \text{ mm}$ ) where UCN with energies exceeding  $h$  were removed during the cleaning time  $t_c \approx 100\text{--}200$  s. It takes  $\approx 100$  s to achieve a sufficiently narrow width ( $\lesssim 2$  mm) for the upper cutoff, since UCN with energies slightly exceeding the absorber height are removed only if they travel within a narrow angular range about the vertical direction.  $t_c$  was increased also by the wobble of the disk by 2 mm. Thus the shortest measurable storage times were about 100 s.

The UCN remaining in the trap after storage were discharged and counted during 200 s (about 4 times the time constant for emptying). After waiting further 100 s, background was reached and counted for 100 s; then the cycle was repeated with variable storage times and scanning heights. For consistency checks, background levels were also measured during the filling and storage phases. The results were generally the same; see [6] for details. The sequence of shutter and stepping motor operations required for system 1 was controlled by a CAMAC/PC system.

In the experiments using both systems 1 and 2, monochromatic UCN were continuously filled into the trap of system 2 during 200 s, with both shutters and the trap open. The spectral width was set to  $h_0 = 9$  mm, larger than for the measurements in system 1 in order to collect about the same number of UCN per trap filling ( $\approx 10^2$ ) in spite of an intensity loss due to unavoidable gaps in the transfer guide system. During filling, the roof in system 2 was at 130 mm above floor. This level approximately corresponded to the UCN gas surface in system 1, taking into account its lift by about 15 mm due to Doppler shift at the trap floor when it is tilted to the horizontal position at the end of filling [7].

During the initial 30–50 s of storage the absorber roof was kept at 130 mm for spectral “pre-cleaning”. Pre-cleaning was used to sharpen the upper edge of the spectrum. Then the absorber was lifted to  $h_s = 135$  mm. The final 50–100 s of storage were used for scanning and spectral cleaning at height  $h$  ( $100 \leq h \leq 135$  mm). For each setting of  $h$ , essentially only those UCN survived this cleaning process, whose maximum jump height was less than  $h$ . In this way the integral spectrum was measured, *i.e.*, the spectrum of all UCN with energies up to  $h$ . The remaining UCN were then channeled to the detector, using the same sequence of counting, waiting and background measurement as for system 1.

The servo motors used for tilting the bottom plate and positioning the roof with a verified precision of  $\approx 0.01$  mm were controlled by a microprocessor unit synchronized with the CAMAC system.

### 4. Data and Data Fitting

Figure 2 shows the data for a measurement with the combined systems. Each point  $N(h_i)$  represents the mean UCN counts per trap filling, obtained from five complete cycles where each cycle consisted of a pseudo-random sequence of 5 storage times  $t_s$  (from 100 to 800 s) and 10 scanning heights  $h$  (from 102 to 135 mm). Clearly visible are the following features: background in the region  $h \leq 108$  mm (1.9 counts during the 200 s of signal counting), a ramp in the range  $114 < h < 129$  mm to which the UCN spectrum  $dN/dh$  is confined, and an asymptotic region ( $h > 130$  mm) above the highest UCN energy in the trap. The asymptotic

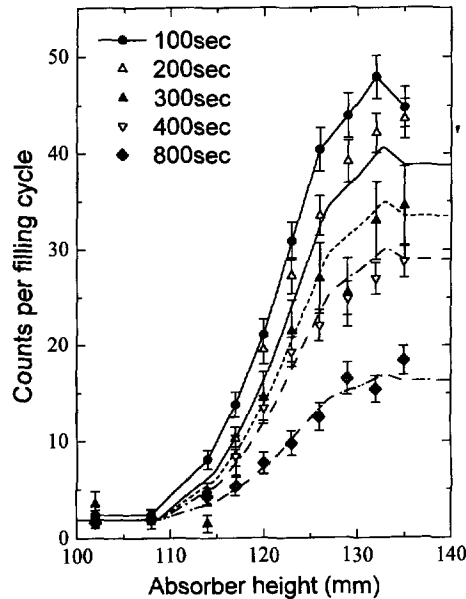


Fig. 2 — UCN spectra measured in system 2 as a function of absorber height  $h$  after various storage times  $t_s$ .  $h$  is measured from the trap bottom level. The fitted curves are explained in the text. In this run the gap between fence top and monochromator absorber was 9.0 mm, and the pre-cleaning and cleaning times were 30 and 50 s, with the absorber at heights 120 mm and  $h$ , respectively. During the remaining time of storage, the absorber was at 135 mm (above the neutrons' "reach")

value  $N_{\text{tot}}$  (background subtracted) decreases with storage time with a characteristic time constant  $\tau \approx 600$  s (this implies that imperfect wall reflections account for only 40% of the loss due to neutron  $\beta$ -decay with its mean lifetime of 888 s). A quick calculation of mean spectral height  $\langle h \rangle = \int h dN/N_{\text{tot}}$  shows noticeable shifts (with  $\approx 1\sigma$  confidence level), namely by  $+(0.5-1)$  mm toward spectral heating, both between the data for  $t_s = 100$  s and for 200 s, and between 200 and 400 s (but apparently not afterwards). The data for a measurement with minimum storage time 250 s shows the same heating effect between 250 and 450 but no statistically relevant change between 450 and 750 s. If such an energy shift is genuine, it is extremely surprising. Searching for a simple explanation we have, therefore, performed a detailed analysis of expected spectral evolution based on the following model:

(a) We take as the initial spectrum  $(dN/dh)_i$  the histogram obtained from the data for the shortest storage time (100 s for the data of Fig. 2) by linear interpolation between successive points  $N_i$  ( $i = 1$  to 10), using a fine grid of 500 to 1000 points. The grid covers a somewhat wider  $h$ -range than the data points to allow simulation of possible spectral blurring and/or shift during the course of storage. The statistical errors of the initial data points are disregarded at this stage (they are taken into account in step (e) below).

(b) This initial spectrum is subjected to temporal exponential decrease,  $\propto \exp(-t/\tau_s)$ , characterized by the storage lifetime  $\tau_s$  which is determined by fitting. On the basis of elementary wall reflection theory,  $\tau_s$  can be assumed constant within the narrow range of UCN energies stored in system 2. (The expected slight decrease with increasing  $h$  was simulated in fits varying also the slope  $d\tau_s/dh$  but this gave a negative slope compatible with zero and no reduction of  $\chi^2$ . Moreover, a negative slope would not simulate spectral heating but cooling, since faster UCN would be lost sooner)

However, in the experiments using system 1 only, the losses do depend on  $h$ , even within the narrow spectral range stored in chamber II, since UCN can disappear by a further loss process: by jumping over the fence back into chamber I (which is drained of UCN soon after closing the entrance shutter, due to high reflection losses on the untreated walls). Therefore, in the fits of spectra for system 1 we used two asymptotic storage lifetimes for chamber II:  $\tau_{sh}$  for energies well above the fence top, where the two chambers are well coupled (fits gave  $\tau_{sh} \approx 100$  s), and  $\tau_{s1}$  ( $\approx 400$  s) for  $h \leq 0$  where the chambers are isolated. An analysis for the transition region yields two exponential decay constants: a fast decay describing transient phenomena and a slow decay describing the long-time behaviour. Since our storage times were sufficiently long the relevant storage lifetime  $\tau_s(h)$  in the intermediary region of  $h$  is given by the longer time constant.

(c) The repeated reflections on vibrating trap walls lead to spectral Doppler broadening characterized by a Gaussian standard deviation  $\sigma_d$ . In addition, a kinematical factor slightly favouring reflections on approaching wall elements over those on a receding wall leads to an increase of mean energy  $\bar{\varepsilon}$  ( $= 173$  mm for system 1) by [8]  $\delta\bar{\varepsilon} = f\sigma_d^2/\bar{\varepsilon}$ , where  $f = 1$  for the symmetrical trap geometry considered in [8], and  $f = 0.88$  for the trap in system 1. For our data, the vibrational broadening during 100 s of storage was  $\sigma_d = (0.7 \pm 0.2)$  mm (fitted result) for a measurement in system 1 without vibration damping, and much less for all other data. The associated shift  $\delta\bar{\varepsilon} < 0.02$  mm is much smaller than observed shifts (30 times smaller in the worst case and completely negligible for the data with vibration damping).

(d) Guided by the much larger measured energy shifts observed in all our data we have included fitting parameters for up to three shifts for different temporal phases of storage. "Early" and "later" shifts were distinguished in one of two ways: either directly on the basis of time elapsed since trap filling (first time bin between 100 and 200 s, second bin between 200 and 300 s, *etc.*); or on the basis of number of wall reflections undergone since the beginning of storage, taking into account that about 15 reflections per second take place in system 1 but only 6.5 per second in system 2. This latter scheme allows a direct comparison of all data for the two systems with the prediction of [4] if we include a further scaling of shifts (by  $\approx 4\%$ ) taking into account that the speculative mean spectral change per wall collision is proportional to  $\langle v_z^2 \rangle^{1/4}$  [4], where the squared vertical UCN velocity is averaged over all wall collisions, with  $\langle v_z^2 \rangle = 1.32 \text{ m}^2\text{s}^{-2}$  for system 1 and  $\langle v_z^2 \rangle = 1.11 \text{ m}^2\text{s}^{-2}$  for system 2. Finally, the measured storage time intervals in the two systems do not correspond exactly to common intervals of reflection number. Scaling was required to obtain consistency.

(e) Using this model of spectral evolution a least-squares fitting routine yielded optimal values for the various parameters (storage times, broadening, shifts; whichever were floated) and their statistical uncertainties due to the statistical errors of all count rates *except* the initial data (for the shortest storage time). The effect of the initial uncertainties was determined by running the program many times with initial data randomized in accordance with a Gaussian probability distribution centred at the measured points and with a width given by their standard deviation. The resulting fluctuation of parameters was added to the errors (squared).

## 5. Results

We have applied this scheme to the data of Figure 2, varying only  $\tau_s$  and  $\sigma_d$ , while all shifts were set equal to zero. This corresponds to the "natural expectation" based on the fact that the system is conservative apart from very small effects of vibrations, and of thermal inelastic scattering at the wall molecules. The latter can, however, induce only large energy transfer (compared to the scale  $10^{-10}$  eV of energy changes considered here) and, therefore, only UCN losses which are already contained in  $\tau_s$ .

Under these assumptions, a best fit, with  $\chi^2 = 1.83/\text{degree of freedom}$ , is obtained for  $\tau_s = 581 \pm 35$  s and  $\sigma_d = 0$  (within the error of 0.2 mm; henceforth  $\sigma_d$  was set equal to zero for this data set without affecting the quality of fits). The fitted curves show pronounced deviations from the data points in the high- $h$  region, which is obviously “sacrificed” for fair representation in the ramp region. A significant reduction of  $\chi^2$  to 1.52, and a much better data representation over the entire  $h$ -range, was obtained when up to 2 early shifts were also considered variable, yielding  $s_1 = 0.6 \pm 0.6$  mm for the shift between 100 and 200 s,  $s_2 = 0.8 \pm 0.6$  mm for the total shift between 200 and 400 s (assuming two equal halves between 200 and 300 s, and between 300 and 400 s), and  $\tau_s = 642 \pm 43$  s. To test the “fragility” of these shifts (each within about one standard deviation) we ran a fit where only  $s_1$  and  $s_2$  were varied, leaving  $\tau_s$  fixed at 581 s, the value optimized for  $s_1 = s_2 = 0$ . This gave the same trend to positive shifts, namely  $s_1 = 0.5 \pm 0.7$  mm and  $s_2 = 0.5 \pm 0.5$  mm with a noticeable decrease of  $\chi^2$  from 1.83 to 1.63, *i.e.*, the data indicate finite, positive shifts independently of the precise value of  $\tau_s$  (and, as further studies mentioned in Section 4, (b), showed, also if an adjustable variation of  $\tau_s$  with  $h$  is included as a further degree of freedom) [9].

These results were obtained assuming that no further spectral change occurred after 400 s of storage. Adding a third possible shift did not improve  $\chi^2$ . Thus, the data of Figure 2 were fitted well with two finite spectral shifts occurring during the early stage of storage. Both shifts are positive and at the  $1\sigma$  confidence level. This implies that, statistically, the probability for both shifts to be compatible with zero (the expectation simply based on energy conservation) is only  $\approx 5\%$ .

A further data set was taken on the combined systems 1 and 2, using storage times 250, 450, and 750 s and longer pre-cleaning and cleaning times than for the data of Figure 2, namely 50 and 100 s, respectively. Five data sets were measured using system 1, under various conditions of vibration isolation, of initial absorber gap width ( $h_0 = 3$  mm or 1.5 mm), for initial storage times of 110 s or 210 s (with 110 or 210 s also used for spectral cleaning), and successive storage times up to 810 s. Figure 3 shows one data set measured on system 1 with  $h_0 = 3$  mm and storage times 110, 210, ..., 610, 710 s.

Each of these sets of data was fitted using the scheme outlined earlier, with due account to the additional loss rate for the system 1 data due to “back-jumping” [10] and, for the data taken with poor vibration isolation, due to vibrational broadening. All the fitted shifts were consistent with the values given above for the data of Figure 2. Consistency between the storage lifetime parameters  $\tau_{sh}$  and  $\tau_{s1}$  for all data of system 1 was also established

Therefore, we performed a common fit to all seven data sets, using the appropriate initial spectrum for each time-evolved spectrum, and adjusting jointly the parameters common to a part or all of the data. Due to the collective analysis, the precision with which the common parameters can be determined is significantly improved since they are determined by all data sets sensitive to a particular parameter, not only by one. In this way, a best fit to a total of 355 data points, evolving from a total of 75 initial spectral points, was obtained by optimizing 7 parameters simultaneously:  $\tau_{sh}$  and  $\tau_{s1}$  for the measurements on system 1;  $\tau_1$  and  $\tau_2$  for the two measurements with the combined system; and three shifts:  $s_I$  for the first 650 wall reflections after 100 s of storage in the combined system, as well as  $s_{II}$  and  $s_{III}$  for the next and consecutive 1500 wall reflections in system 2. Where necessary, the shifts were scaled according to the scheme outlined in Section 4, (d).

The results were:  $\chi^2 = 1.76$  for  $\tau_{sh} = 104 \pm 40$  s,  $\tau_{s1} = 450 \pm 40$  s,  $\tau_1 = 634 \pm 42$  s (storage lifetime for the data of Fig. 2),  $\tau_2 = 486 \pm 30$  s,  $s_I = 0.76 \pm 0.46$  mm,  $s_{II} = 0.50 \pm 0.17$  mm, and  $s_{III} = -0.10 \pm 0.19$  mm. The spectra calculated with these parameters for the data of Figures 2 and 3 are presented as curves.

These results for spectral shifts are plotted in Figure 4 in the form of shift per 1000 wall



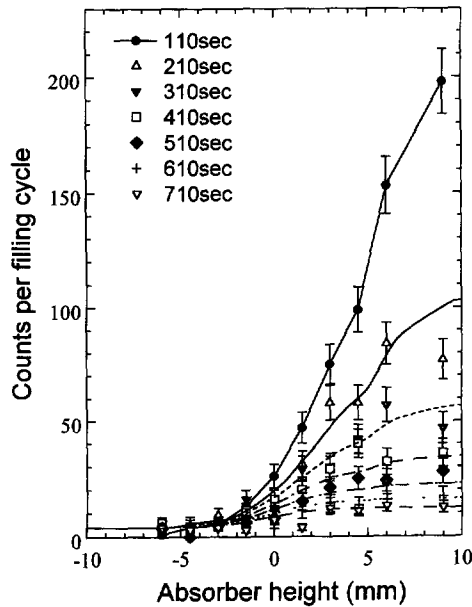


Fig. 3. — UCN spectra measured in system 1 as a function of absorber height  $h$  above fence top level after various storage times  $t_s$ . Negative values of  $h$  are possible since the disk had a circular groove at the fence radius, allowing its active surface to be lowered below the fence top level by 9 mm. Here, the initial gap  $h_0$  between fence top and absorber was 3.0 mm, and the cleaning time was 110 s (no pre-cleaning used), with the absorber at height  $h$ . During the remaining time of storage and during emptying, the absorber was at  $h = 9$  mm (above the neutrons' reach).

reflections. The initial interval of 400 reflections is an estimate of reflections taking place in trap 2 before the first energy scan was performed. Due to the long duration of scanning (50 s, and longer in other cases) the “storage time before the first scan” cannot be precisely specified while the time intervals between successive spectral scans are well defined.

The surprising result is: Two positive “early shifts” were established with a reliability of respectively 1.7 and 3 standard deviations. No finite third shift ( $s_{III}$ ) was observed within its  $1\sigma$  error margins, nor were any later spectral shifts found. (They were all set equal to zero without impairing the quality of fit). Combination of shifts on the basis of storage time gave the same trend (one positive shift of 0.5 mm between 100 and 200 s, at the  $3\sigma$  level; possible later shifts compatible with zero).

Looking for systematic errors, we have tried to eliminate all possible uncertainties in the course of this experiment: *Wall vibrations* and storage losses due to *back-jumping* in system 1 are difficult to analyze and have, therefore, been eliminated in our second set of measurements using the combined system. This did not change the observed spectral shifts. The *cut-off characteristics of the absorbers* are not exactly known but the spectral cleaning times for given value of  $h$  were chosen identical for different storage times in a given data set, taking into account the time needed for vertical movement and making sure that residual small differences of cleaning conditions would tend to simulate, not the spectral heating indicated by our data, but cooling. Therefore, our relative measurements of spectra after different storage times are unaffected by details of absorber characteristic. In the analysis we assumed *isotropy* of UCN flight directions. In principle, a preference of horizontal over vertical direction, sustained over

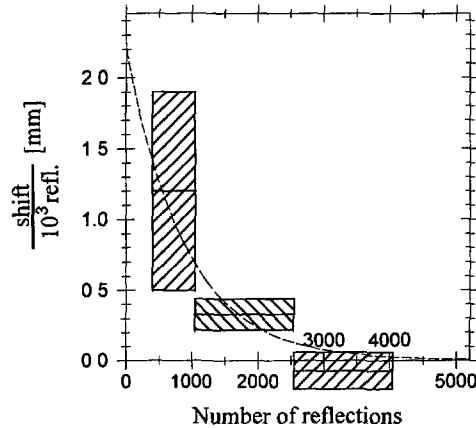


Fig. 4. — Temporal evolution of UCN spectra, shown as shifts per 1000 wall reflections in the traps and plotted *vs.* the number of preceding wall reflections. The width of the shaded regions indicates plus/minus one standard deviation. The dashed exponential curve is a guide to the eye, showing the total shift of about 2 mm (the area under the dashed curve) and the decay constant of about 900 collisions.

times of order of the storage times ( $\geq 100$  s), could simulate a spectral change. But the Fomblin grease applied to the traps' side walls (and in trap 2 also to the bottom plate) made the surfaces very rough (optically dull; hence the estimated roughness of  $> 1 \mu\text{m}$  is much larger than the UCN wavelength of  $\approx 10^2$  nm). Thus the directional relaxation times are at most of order 1 s (*i.e.*,  $\ll 100$  s). Finally, how about heating by *micro-Doppler shift* on the rotating absorber lid? This cannot apply to system 1 where only the external rim of the absorber disk, which moves fast, is “in contact” with the stored UCN. It could apply to the centre region of the absorber in system 2 but, due to a technical problem, all our measurements with trap 2 were performed with absorber at rest. *In summary*, we have not found any systematic effect which might simulate the observed initial micro-heating of stored UCN, nor is this phenomenon understandable on the basis of energy conservation. Our data are also incompatible with the indication of fairly large spectral UCN cooling in [2] (which, however, was not claimed).

Yet, the possibility of very small initial spectral change is contained in the speculative model described in [4] and [11], and therefore we now compare our results with this prediction.

## 6. Comparison with an Unconventional Model

Motivated by the age-old problem whether purely exponential decay of an unstable system should exist or not we suggested in [4, 11] that the puzzle may be resolved in favour of exponential behaviour by introducing a novel degree of freedom related globally to any interaction. In this view, the ordinary effects of interactions are supplemented by the property to induce statistical changes of state. For example, if a slow neutron moves in a gravitational field, the change can be described as scattering by the field from a state of free flight in one direction at a certain velocity to a state with slightly different parameters. The uncertainty relation was used to determine the smallest observable change in such a transition, thus establishing a rule for breaking the flight parabola up into short segments of “free flight”. This defines a short mean lifetime  $\tau = (2gp_z/\hbar)^{-1/2}$  [4] of the free, plane-wave state, as well as an associated energy scale  $b = \hbar/2\tau$ . Here,  $g = 9.8 \text{ m s}^{-2}$  on the earth's surface and  $p_z$  is the magnitude of

vertical momentum component. Are there measurable manifestations of  $b$ ? UCN, for which  $b \approx 10^{-11}$  eV, represent a singular testing field for these ideas since  $b$  is not hopelessly small compared with the typical UCN energies of  $10^{-8}$  to  $10^{-7}$  eV.

In the concrete model [4, 11],  $b$  is used to scale the non-linear term of a "logarithmic Schrödinger equation" (LSE) of the type first suggested by Białynicki-Birula and Mycielski [12]. The model [11] develops the LSE in terms of probability concepts which in turn require additional variables  $\mathbf{x}_\tau$  attributed to the postulated novel degree of freedom, in the following form

$$i\hbar[\partial\psi_\tau(\mathbf{x}_\tau, t)/\partial t] = [-(\hbar^2/2M)\nabla_\tau^2 - b \ln(a^n|\psi_\tau|^2)]\psi_\tau(\mathbf{x}_\tau, t), \quad (1)$$

where  $M$  is the mass,  $n$  is the space dimensionality,  $\nabla_\tau = \partial/\partial\mathbf{x}_\tau$ , and  $a$  is a non-observable length. In the interpretation [11],  $\mathbf{x}_\tau$  denotes a relative coordinate between the particle position  $\mathbf{x}_c$  and its mean value  $\mathbf{x}$  taken over the unconventional probability distribution  $|\psi_\tau|^2$ .  $\psi_\tau$  is different from the ordinary wave function  $\psi$ , which in our model satisfies the usual, linear Schrödinger equation for ordinary space. A further difference is the absence of a potential  $V$  in equation (1). In this model,  $V$  appears in the Schrödinger equation for  $\psi$  but in equation (1) this term is replaced by  $\mathbf{x}_\tau \cdot \nabla V(\mathbf{x})$  [11]. This term is omitted in equation (1) since it contributes little when  $V$  varies slowly on the scale  $L$ , where  $L = \hbar/\sqrt{2Mb} = \sqrt{\hbar\tau/M}$  is of order  $\mu\text{m}$  for UCN.

A variety of stationary, normalized solutions of form  $\psi_\tau \propto \exp(-i\omega_\tau t)$ , with constant  $\omega_\tau$ , have been obtained in [4, 11, 12]. The definition of their energy in the form

$$E_\tau = \langle -(\hbar^2/2M)\nabla_\tau^2 \rangle - b \langle \ln(a^n|\psi_\tau|^2) \rangle \quad (2)$$

[4, 11] follows directly the scheme outlined originally in [12] and illustrates the role of  $b$  as the scaling factor for energy [13]. The averaging symbol  $\langle \rangle$  denotes the expectation values  $\langle \psi_\tau | \dots | \psi_\tau \rangle / \langle \psi_\tau | \psi_\tau \rangle$ . As shown in [12], the spectrum of  $E_\tau$  defined by (2) is bounded from below for the "gausson"  $\psi_\tau \propto \exp(-x_\tau^2/2L^2)$  [12]. The first term in equation (2) resembles an internal energy and is a constant,  $nb/2$  [12], for all stationary states. Therefore, the second term (which "has the form of an entropy" [12]) determines the energy. For states where  $\psi_\tau$  is spread out over a large volume  $\Omega$  in  $\mathbf{x}_\tau$ -space, the mean value of  $|\psi_\tau|^2$  is small ( $\approx \Omega^{-1}$ ), and therefore  $E_\tau$  increases with  $\Omega$  roughly as

$$E_\tau \approx b \ln(\Omega/L^n), \quad (3)$$

and  $E_\tau = 0$  is a convenient choice for the gausson ground state [12]. Evaluation of  $\Omega$  from values of  $\langle x_\tau^2 \rangle$  for a number of numerically calculated stationary solutions  $\psi_\tau$  shows that the estimate in equation (3) is valid within a maximum error of  $\approx b \ln 2$ .

It was suggested in [4] that for UCN stored in a material trap transitions to different  $\psi_\tau$ -states can take place, not during free flight, but in wall reflections where the wall ensures the conservation of momentum (linear and angular) needed to make swapping of energy between  $E_\tau$  and the measurable energy  $E$  for the ordinary degrees of freedom ( $E = T + V$  with  $T = Mv^2/2$  and  $V = Mgz$ , where  $v$  is the velocity and  $z$  the height) possible under the constraint of conservation of total energy  $E_t = E + E_\tau$ .

The prediction of [4] is a random walk in  $E$ - and  $E_\tau$ -space, starting from  $E_\tau \approx 0$  and leading to slight spectral blurring and a net initial micro-cooling by about 3 mm during the initial few thousand wall collisions in the trap (the latter is a very rough estimate). The asymptotic energy change of 3 mm was calculated from (3) for  $b = 10^{-11}$  eV and trap dimensions  $D \approx 10$  cm so that the maximum volume  $\Omega_{\text{max}}$  in  $\mathbf{x}_\tau$ - (and  $\mathbf{x}$ -) space is  $D^3 \approx 10^3$  cm<sup>3</sup>.

A comparison with the data of Figure 4 shows consistency with the magnitude and speed of the predicted energy change *but the opposite sign* (heating rather than cooling). The area under the dashed curve in Figure 4 (an exponential curve drawn as a guide to the eye) is the total shift  $s_{\text{tot}} \approx 2$  mm. This checks with the expectation  $s_{\text{tot}} = 2.2$  mm based on (3) for  $b = 6 \times 10^{-12}$  eV (for  $\langle v_z^2 \rangle = \langle p_z^2 \rangle / M^2 = 1.11 \text{ m}^2\text{s}^{-2}$ ),  $L = 1.9 \text{ } \mu\text{m}$ , and  $\Omega_{\text{max}} \approx 0.04 \text{ m}^3$  for the volume occupied by the UCN gas in trap 2. The measured time constant for slowing down to  $1/e$  of the initial shift/reflection corresponds to about 900 reflections which is of the predicted order of magnitude [4]. (This estimate was based on a perturbation approach to calculating transition probabilities between  $\psi_\tau$ -states, but suffers from the lack of analytical solutions to equation (1). Approximations to only a certain class of stationary solutions were used and other classes were neglected. Since the measured speed of spectral change is 3-5 times higher than expected it appears that all classes of solutions have to be considered in an improved analysis.)

But the sign of the shift is “wrong”. Since the measured errors for the initial shifts exclude compatibility with zero with probability  $> 99\%$ , they exclude the opposite signs with even greater certainty. Can the model be adjusted to invert the sign of  $E_\tau$  without changing the magnitude and its temporal evolution?

This would, obviously, be achieved by inversion of the sign of the log-term in equation (2). This proposition requires two remarks: (a) The negative sign of the log-term in equation (1) is required to obtain normalizable solutions. (b) However, the sign of the log-term in an expression for measurable energy within a new concept is not uniquely defined, but the arguments can only draw on analogies (and nature decides). The analogy is: “Energy” involving a log-term may have the form of a free energy which is given by  $U - TS$  in thermodynamics, where  $U$ ,  $T$  and  $S$  are respectively the internal energy, temperature and entropy. Suppose we associate with  $S$  the entropy of information,  $S = - \sum_i P_i \ln(P_i)$  for the probabilities  $P_i$  of mutually exclusive events, identify  $P_i$  with the probability distribution  $|\psi_\tau|^2$ , and replace the thermal energy  $k_B T$  (with Boltzmann’s constant  $k_B$ ), which scales the entropy term in thermodynamics, by  $b$  (as suggested in [11]). Then we obtain equation (2) *except* that the sign of the log-term becomes positive (as was also noted in [11]). The irrelevant constant length  $a$  is needed only to ensure correct dimensionality. Thus the re-defined energy is given by

$$E'_\tau = -(\hbar^2/2M)\nabla_\tau^2 + b \langle \ln(a^n |\psi_\tau|^2) \rangle, \quad (4)$$

where the first term on the right, as in equation (2), is a constant,  $nb/2$ , for all stationary  $\psi_\tau$ -states. In our thermodynamic picture this constant has the same form as the internal energy  $nk_B T/2$  of a monatomic gas (if  $k_B T$  is replaced by  $b$ ). Being constant it is irrelevant for a non-relativistic energy.

By adjusting the length  $a$ , the energy spectrum for the new sign convention can be chosen identical to the old one, *apart from the sign*. Thus,  $E'_\tau = -E_\tau$  for any stationary solution to equation (1) (and this choice is possible for any norm of  $\psi_\tau$ ). As a consequence, the lower bound ( $= 0$ ) for  $E_\tau$  changes to an upper bound to  $E'_\tau$ , and one may then worry about the stability of such a model (can the energy “crash to  $-\infty$ ”?). However, in all practical cases the spectrum is bounded also from below due to the finite value of available volume  $\Omega_{\text{max}}$ . (Even taking, unrealistically, for  $\Omega$  the volume of the universe instead of that of trap 2, increases  $\ln(\Omega/L^3)$  in equation (3) only by the factor 6, and  $|E'_\tau|_{\text{max}}$  would be only  $\approx 10^{-9}$  eV.)

Summarizing, the only effect of the change of sign for the log-contribution to energy is: the initial micro-cooling prediction changes to initial micro-heating for UCN stored in a trap with fixed walls, leaving the magnitude and speed of spectral change the same since they are not affected by the sign [4]. Since the modified model is consistent with our data, while the original

one is not, it seems fair to state that we have measured the positive (not negative) sign for the new log-contribution to total energy  $E'_t = E + E'_\tau = E - E_\tau$ , which is conserved throughout storage. The development of  $\psi_\tau$  during storage can be viewed as expansion in  $\mathbf{x}_\tau$ -space, from volume  $\approx L^3$  to the trap volume, this expansion reducing  $E'_\tau$  exactly as for the free energy of an ideal gas during isothermal expansion

## 7. Conclusion

The UCN experiments described gave a clear indication of a slight initial micro-heating for UCN stored in a trap. The effect cannot be explained by wall vibrations or energy dependence of wall losses and it even seems to contradict energy conservation. However, it is consistent with an unconventional model [4] which had predicted the observed magnitude and temporal evolution of spectral change without invoking violation of any established conservation law. Yet the sign of the observed change (heating, not cooling) required a modification to the model which appears plausible. Since the model is rooted on the old problem of quantum mechanics, whether the decay of an unstable system (like atoms in an excited state, or particles subject to  $\beta$ -decay) should proceed purely exponentially or not, we interpret the data as providing evidence for a strictly exponential decay law. The experiments were indirect but much simpler than direct observation of the long-time decay pattern (which has so far also failed to find evidence for any deviation from the exponential law at long times).

The spectral change observed is smaller and of opposite sign than the indication in [2] which the authors mentioned but dismissed. The new data and our model are consistent with the broadening by  $\approx 10^{-11}$  eV of a grating diffraction line for UCN reported in [3] (in this case the change of sign for energy is irrelevant for the broadening by  $\approx b = 1.2 \times 10^{-11}$  eV [4]). Initial micro-heating of a UCN gas would tend to cause losses in a storage experiment but appears too small to explain the "anomaly" reported in [1]. However, a meaningful comparison with [4, 11] requires a full analysis of the coupling of  $\psi$  and  $\psi_\tau$  due to the presence of the wall potential. For an intriguing observation independent of the shifts but possibly relevant to the "anomaly" reported in [1] see [6].

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- [6] In measurements with system 1 we deduced the background from the data at the lowest  $h$ -values, at least 4.5 mm below fence-top level. This background was slightly higher than the value measured, consistently, during the filling and storage phases and after emptying. Suspecting possible leaks through the fence we found none. (All edges were tightly sealed with Fomblin oil.) While this unexplained circumstance is irrelevant for the spectral shifts reported here it may indicate unexpected transmission through the thick (0.1 mm) Al walls of the fence and thus provide a hint as to the enhanced “anomalous” loss mechanism [1]. This possibility (as well as the shifts) requires further investigation.
- [7] Although the Doppler shift per reflection on the moving bottom plate depends on the position of impact, the associated spectral broadening is small since the successive shifts are averaged out during the many ( $\approx 25$ ) reflections occurring during closing.
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- [9] This test confirms that  $\tau_s$  and the shifts are only weakly correlated (the magnitude of the correlation coefficients  $C_{\tau_s}$  is  $\lesssim 0.3$ ). The two shifts are mutually not strongly correlated, either (with  $|C_{ss}| \approx 0.7$ ), thus separately derivable from the data. A high degree of correlation would result in large parameter errors. These errors are calculated using a parabolic approximation of  $\chi^2$  near its minimum, and the approximation is justified since the actual values of  $\chi^2$  at the extreme points of the ellipsoid of confidence agree with the parabolic extrapolations within  $\approx 30\%$ . As usual, systematic uncertainties increasing  $\chi^2$  were accounted for approximately by multiplying the purely statistical errors by  $\sqrt{\chi^2}$ .
- [10] The back-jumping probability for a spectrum scanned at absorber height  $h$  is subject to uncertainty because the spectral cut-off property of the absorber is not exactly known. The effective cut-off energy is higher than  $h$ , and therefore the count rates in Figure 3 (and for the other 4 data sets for system 1) do not drop to zero abruptly at  $h = 0$  but rather near  $-2$  mm. However, the deviation depends on experimental details and is difficult to measure or calculate precisely. Therefore we tested the stability of fitted parameters against possible variations of effective spectral cut-off energy but obtained only minor variations of the fitted shifts. The full bandwidth is contained in the quoted error margins.
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- [13] The proportionality of energy  $E_\tau$  to  $b = \hbar/2\tau$ , where  $\tau$  is the mean lifetime of a state, has an important consequence. If a beam is split coherently into equal or unequal parts in an interferometer, the probability for the decay to take place in one or the other partial beam scales with the proportion of beam intensity in the two beams. Therefore the  $b$ -values for the beams scale in the same way and add up to the same value as for the initial beam. Thus we expect no “energy of splitting”, and hence no phase shift in an interferometer [14]. Absence of splitting energy conflicts with the ideas of [12, 15] where  $b$  was considered an unknown but universal constant which could be measured in interferometry or diffractive wave dispersion. No measurable shift has been found in thermal neutron interferometry [16a] and in cold neutron diffraction on a straight edge [16b] so far.

- [14] Absence of an interferometer shift persists in a proposed relativistic extension of energy where the rest energy  $E_0 = mc^2$  of a particle is slightly modified to  $mc^2 = Mc^2 - b - E_\tau$  (in the limit  $b \ll Mc^2$ ). This form implies a “bias mass” of order  $b$  (and  $= -b$  for  $E_\tau = 0$ ), which can explain a particle physics “coincidence”: the mass difference  $\Delta m = m_L - m_S$  between the  $K_L^0$  and  $K_S^0$ -mesons is almost exactly given in terms of the lifetimes  $\tau_L$  and  $\tau_S$ , by  $c^2\Delta m = \hbar(\tau_S^{-1} - \tau_L^{-1})/2 = b_S - b_L$  which makes  $K_L^0$  and  $K_S^0$  degenerate in M. This fits into the common view of  $K_L^0$  and  $K_S^0$  as the same particle in different, coherently superposable states. In the language of debated CP violation for the  $K^0$  system the above  $\Delta m - \tau$  relationship translates into the value  $\Phi = 45^\circ$  for the phase of the CP violating amplitude  $\varepsilon$ , very close to the measured value  $\Phi_{00} \approx \Phi_{+-} = 46.5 \pm 1.2^\circ$  [17] (for  $K_L^0$  decay into two uncharged or charged pions, respectively). For a UCN implication see [18].
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- [18] For UCN in a trap, the slight dependence of mass,  $mc^2 = Mc^2 - b - E_\tau$ , on  $b = (\hbar g p_z / 2)^{1/2}$ , and thus on  $p_z$ , can be taken into account in an analysis of the classical flight trajectory between wall reflections. The result is a negligible deviation (by  $\approx b/Mc^2 \approx 10^{-20}$ , relatively) from the usual flight parabola and a justification for our measurement of  $E_\tau$  via a vertical displacement of UCN gas surface, using the usual relationship between energy  $E$  and maximum rise height.