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Experimental study of quasi-elastic scattering of ultracold neutrons

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Abstract. Ultracold neutrons (UCN) are lost from traps if they are quasi-elastically scattered from the wall with an energy gain sufficient to exceed the Fermi potential for the wall. Possible mechanisms of a quasi-elastic energy transfer are, for instance, scattering from hydrogen diffusing in an impurity surface layer or on surface waves at a liquid wall. Using two different experimental methods at the UCN source of the Institut Laue-Langevin we have investigated both the energy-gain and the energy-loss side of quasi-elastic UCN scattering on Fomblin grease coated walls. For Fomblin oil and similar new types of oil we report up-scattering data as a function of temperature and energy transfer. These low-temperature oils may be used in an improved measurement of the neutron lifetime, which requires extremely low wall reflection losses.

PACS. 61.12.-q Neutron diffraction and scattering – 61.12.Ex Neutron scattering techniques (including small-angle scattering) – 66.10.Cb Diffusion and thermal diffusion

1 Introduction

Ultracold neutrons experience total external reflection from the walls of closed cavities and thus can be trapped for long times of the order of the neutron lifetime ($\tau_n = 885.8 \pm 0.9$ s [1]). Beside beta-decay the storage lifetime is limited by wall reflection losses due to nuclear capture and thermal inelastic scattering in the thin UCN/wall interaction layer of ~ 10 nm thickness. For weakly absorbing solid materials like Be at low temperature the calculated losses are very small, but measured values have been 2–3 orders of magnitude larger [2]. The lowest losses measured so far for any material have been reported for liquid Fomblin oil near room temperature; the small losses were crucial for the most precise measurements of τ_n to date [3–7]. Recently new per-fluorinated polymers have been developed (*e.g.*, [8]). They are similar to Fomblin but have an ~ 80 K lower solidification temperature. Such oils were included in our study since they are good candidates for materials with even lower UCN reflection loss [9].

Beside the losses due to capture or due to the large energy transfers in thermal inelastic scattering, a UCN will also quickly disappear from the trap if it suffers quasi-elastic scattering with a small energy transfer to a final energy exceeding the wall potential, however slightly this may be. This could be a reason for the "anomalous" loss [2]. First results of our studies of quasi-elastic scattering have been reported in [10–12]. Energy changes of the order of neV had also been observed in [13] and [14], but since only the energy-loss or the energy gain side were measured these changes were described as evidence of "small cooling" [13] or of "Vaporizing UCN" [14]. The data presented here suggest a common origin: quasi-elastic scattering with positive and negative energy transfers. We compare the data with the models of hydrogen diffusion in an impurity layer on a solid [15] and surface waves on a liquid wall [16].

2 Measurement of quasi-elastic down-scattering

The experimental setup shown in Figure 1 and described in detail in [10,17] was especially suitable for studying quasi-elastic down-scattering. A "fence" (containing a meander-shaped aluminum foil) divides the cylindrical

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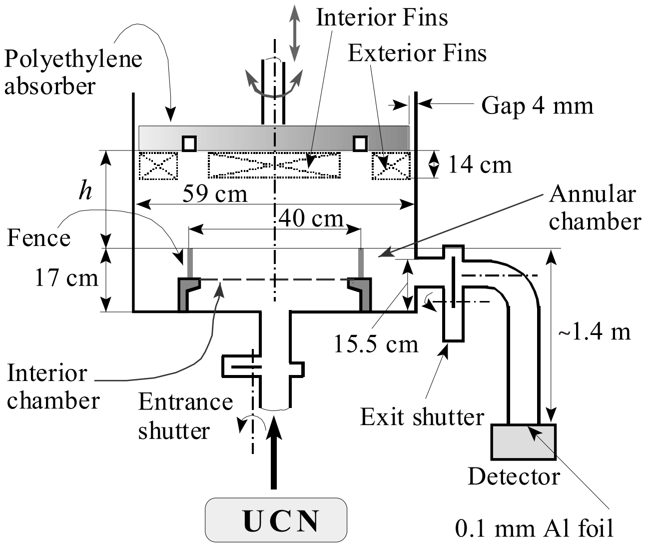

Fig. 1. UCN storage system 1 with Fomblin grease coated walls. UCN in a spectral band are trapped in the annular storage chamber. The low-energy cut-off is determined by the "fence" height, the upper cut-off by the absorber. The absorber was extended downward by polyethylene fins, the exterior extension being optional.

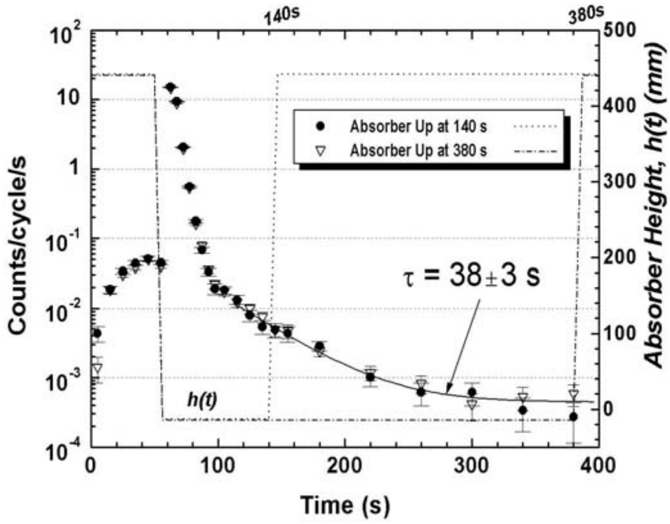

Fig. 2. A measurement cycle: Loading for 50 s with closed exit shutter; then the exit shutter opens and absorber I is lowered to $h_0 = -15$ mm (shown by the dashed lines and the right-hand scale). The data (left scale) show the "long tail" ($\tau \approx 40$ s) and its independence of the time of raising the absorber again (at $t = 140$ s, or much later at 380 s).

vessel into an interior chamber and the exterior, annular storage space for UCN in a well-defined spectral band. UCN enter the center part from below. To reach the Fomblin-grease coated storage volume they must jump over the "fence". This establishes the low-energy cut-off. Placing the Al fence into a Fomblin-oil filled circular groove and coating it with Fomblin grease ensured leak-tightness for all storable UCN. The high-energy cutoff is variable and determined by the vertical position of a rotating "absorber" made of polyethylene (PE). Rotation (at a frequency f that is variable up to 4 Hz) and a corrugated bottom side of the absorber increase its efficiency to reject UCN with jump heights exceeding the absorber height [10].

A circular recess cut into the absorber allows its translation down to a height $h_{min} = -15$ mm *below* the fence top. In this position, the cutoff values from above and below overlap, thus UCN can remain in the annular chamber only if they had lost some energy in quasi-elastic wall collisions before the absorber was moved down. For $h = h_{min}$, the minimum energy transfer is $-mgh_{min} \approx 1.5$ neV since the gravitational potential energy for the neutron is $mg = 1.025$ neV per cm of height.

We studied somewhat larger energy changes using a modification where a fin-like absorber extension was attached to the bottom of the exterior part of the absorber. These four vertical PE plates are indicated in Figure 1 (but not drawn to scale). In the lowest absorber position the fins penetrated 15 cm into the 17 cm high annular chamber. Similar fins were permanently mounted to the interior absorber section to ensure that all UCN were removed from the interior chamber when the absorber was in a low position; thus these UCN could not contribute to the UCN density in the annular chamber. We will refer to the absorber without (with) the exterior extension as absorber I (absorber II).

To ensure a reproducible position of the absorber its motion at a speed of ~ 6 cm/s was controlled with a precision of 0.01 mm. The Fomblin grease layer was several 0.1 mm thick, sufficient for UCN reflection, and the measurements were performed at room temperature in a vacuum of (typically) 5×10^{-5} mbar.

The "long-tail" data

Figure 2 shows the sequence of absorber motions and detector count-rate, averaged over many cycles, for measurements with absorber I (without exterior extension) rotating at $f = 2$ Hz. During trap loading for $t_{fill} = 50$ s the absorber was at a high position, $h_1 = 440$ mm above the fence top, to allow the UCN to fill the entire trap volume. The exit shutter was closed, but the build-up of UCN density is reflected in the increase and saturation of UCN leakage current passing through small gaps of the closed shutter. At the end of loading the entrance shutter closes, the exit shutter opens and the absorber is lowered to $h_0 = -15$ mm, as shown by the dashed lines in Figure 2. Due to the overlap between the upper and lower cutoff energies the UCN are quickly removed from the annular chamber; thus the count-rate drops steeply by a factor > 100 during the interval $60 < t < 90$ s.

However, at $t \approx 100$ s the data show a transition to a slow, nearly exponential decay described by a time constant $\tau = 38 \pm 3$ s. This "long tail" has first been described in [17]. The efflux time constant of $\tau \approx 40$ s is much longer than the maximum value expected if all wall reflections were purely elastic. For our trap geometry, a maximum of 22 s was calculated for the energy of a UCN just barely able to jump over the "fence", and a measurement with $h_0 = +3$ mm (*i.e.*, using UCN energies just slightly above fence top) gave the same result: $\tau = 21.7 \pm 0.5$ s. For these UCN we also measured the storage lifetime τ_s and obtained values in the range $\tau_s = 140 - 200$ s, improving with longer pumping time.

Pulse-height spectra and a comparison of counts measured with narrow and with wider discrimination window settings showed that the long-tail data are due to neutrons, not to $\beta-$ or $\gamma-$ radiation (that would be expected if detector window activation played a role). The "long tail" data in Figure 2 are independent of the time when the absorber is raised again to h_1: at $t = 140$ s or, much later, at 380 s. A further clue to the origin of the slow efflux is provided by a comparison with the data of [17]. The previous value $\tau = 66 \pm 6$ s is consistent with 40 s if we correct for the only change of geometry: In [17] the exit guide cross section had been 1.6 times smaller, namely 38 cm^2 instead of 64 cm^2 in the present experiment. Furthermore, in the later experiments τ increased from 40±3 s to 51±4 s when beryllium foils placed on the storage chamber bottom partly blocked the UCN exit from the annular chamber.

All these data are plausible if we attribute the "long tail" to quasi-elastic energy transfers at the walls. For instance, if UCN with just enough energy to clear the "fence" are down scattered by 4 neV their maximum jump height in the storage chamber will be 2.5 cm below the upper edge of the exit area, which is 15.5 cm above the bottom. Due to the smaller effective exit cross section and the lower mean velocity the efflux time constant is increased to the range of ≈ 40 s.

To determine the energy dependence of quasi-elastic scattering we varied the cleaning height h_0 of absorber II (with extension fins) as indicated in Figure 3, keeping $h_1 =$ 10 mm (position during loading for 170 s) and $h_{emp} =$ 50 mm (position after $t = 240$ s) fixed. No exit shutter was used in these measurements, thus the count-rate measured during loading was high: about one half of the intensity in the primary neutron guide in the same spectral range, as expected since the entrance and exit guide cross sections were the same. In Figure 3 the "long tail" appears for all h_0 down to -70 mm (and even for -100 mm in other runs), with decreasing intensity. It disappears only for the lowest position $h_0 = -152$ mm (absorber 19 mm above the bottom) which is below the lower edge of the exit cross section; thus, UCN with such low energies cannot reach the height of the exit area.

A comparison between Figure 2 and Figure 3 shows a significant difference of efficiency of the two absorbers for a given cleaning height (compare $h_0 = -15$ mm for absorber I with $h_0 = -10$ mm or $h_0 = -30$ mm for absorber II). In contrast with Figure 2 the data for absorber II (Fig. 3) show an increase at $t = 240$ s, when the absorber II is raised after 80 s of cleaning. The bump is due to the few higher-energy UCN still present in the trap at this time. The more efficient absorber I shows no bump.

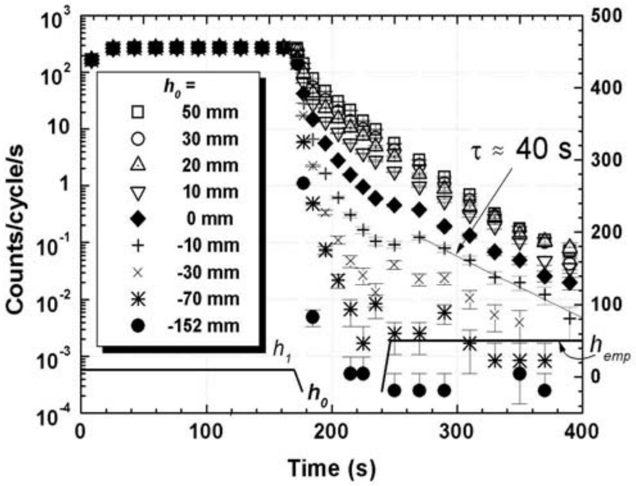

Fig. 3. Time spectra measured as a function of the "cleaning height" h_0 of absorber II. h_0 refers to the bottom edge of the absorber "fins" attached to the exterior part of the absorber. The absorber heights $h_1 = 10$ mm and $h_{emp} = 50$ mm were kept fixed and no exit shutter was used in these runs. The "long tail" appears all the way down to $h_0 \approx -100$ mm.

The efficiencies of both absorbers improved with higher rotation frequency, and we may conclude that polyethylene absorbers used in the past may also have been less efficient than calculated for the clean material, especially if no rotation was used.

The reduced efficiency of absorber II does not affect the main conclusion: the "long tail" disappears only slowly with lower cleaning height. We can relate [10] the integral "long tail" intensity to the net frequency of UCN wall collisions during loading, which was measured with exit shutter removed as for the data of Figure 3. The collision frequency for the entire annular chamber wall area is determined from the count-rate measured during loading, which reflects the flux at the exit area, taking into account the dependence of UCN density from height due to gravity. In converting the data with open exit (Fig. 3) to the value with closed exit shutter (Fig. 2) an uncertainty of a factor ~ 2 arises since the effective constriction of UCN passage over the fence can only be estimated. The result is the estimate $p_{down} \approx 1.4 \times 10^{-6}$ for the mean probability of quasi-elastic down-scattering, per wall collision, by at least ~ 4 neV.

3 Measurement of quasi-elastic up-scattering

We performed these measurements to test the suitability of two new types [18] of "Low Temperature Fomblin" (LTF) for a neutron lifetime experiment. One (here called LTF1) had a molecular weight $M = 4883$ and a vapor pressure $P \leq 7 \times 10^{-5}$ mbar at room temperature (r.t.). For the other (LTF2) the values were $M = 2354$ and $P = 1.5 \times 10^{-3}$ mbar. We used the spectrometer [19] shown in Figure 4, to study the temperature dependence of the energy-gain side of quasi-elastic UCN scattering, comparing the data for LTF1 and LTF2 with Fomblin grease and

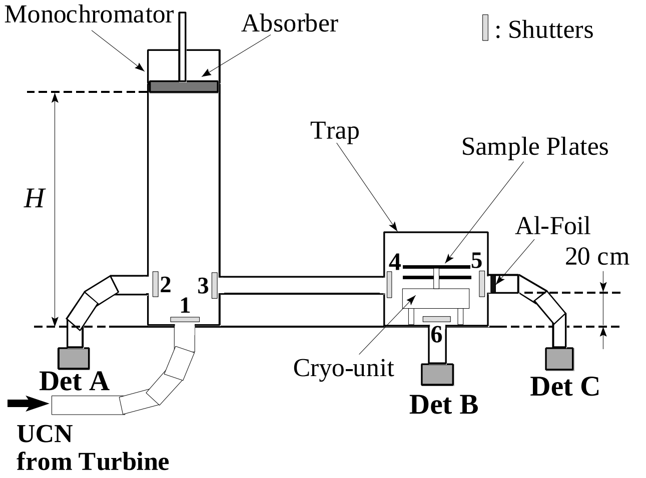

Fig. 4. Setup 2 used to measure quasi-elastic up-scattering on Fomblin grease, on Fomblin oil and on two kinds of "Low Temperature Fomblin". Detector C counts only UCN that penetrate the Al foil after quasi-elastic up-scattering on the samples. 1-6: UCN shutters.

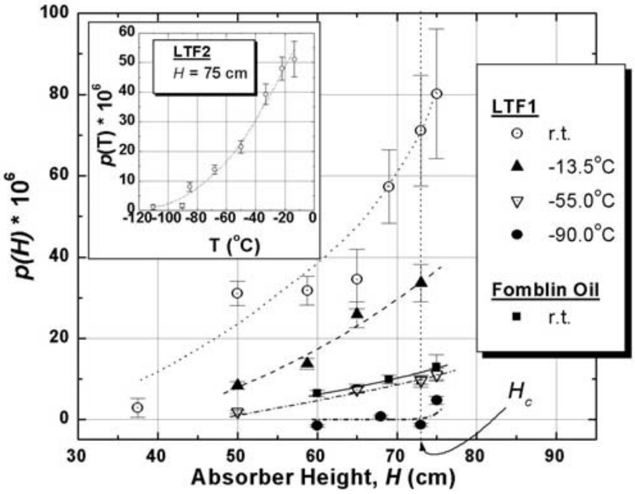

Fig. 5. Quasi-elastic up-scattering probability p for the LTF1 oil. The minimum energy transfer is determined by the difference between the Fermi cutoff for the Al window at $H_c = 73$ cm and the UCN spectral cutoff at the absorber height H. The inset diagram shows the T-dependence of p for LTF2. The curves are guide to eye.

oil at r.t. After filling the Be-coated cylinder, shown on the left, for 100 s, a spectrum with a sharp upper cutoff determined by the absorber height was prepared. Since the losses were small, long cleaning times of 100 or 200 s could be used. The UCN were then transferred to the carbon-coated UCN trap containing a cryogenic unit kept at -110 to $+25$ °C. Two plates were mounted on this unit, and three of their surfaces, with a total area of $\sim$ 40 dm^2, were coated with the sample substance.

Port C of the trap was tightly closed by a 12 μm thick Al-foil blocking all UCN whose kinetic energies, measured at the lowest part of the foil, were less than the Fermi potential $V_c = 54$ neV for Al. If the absorber height H was below the corresponding critical value $H_c = 73$ cm, UCN could be counted by detector C, with open shutter 5, only if they had been quasi-elastically up-scattered on the trap walls by $\Delta E \geq \Delta E_{min} = mg(H_c - H)$. We measured the integral counts of detectors C and B during UCN transfer (for 100 s; with shutters 3, 4, 5 open, 6 closed) and during the subsequent "measuring period" (100–600 s; with 4, 6 closed, 5 open). The leakage through small gaps of the closed shutter 6 to detector B served as a measure of UCN density. These count-rates were combined with calibration and empty-trap background data and with the count-rate of detector B, which was measured during the ensuing period of trap emptying (for 100–150 s; with 4 closed, 5, 6 open) to determine the total UCN number in the trap. The result is the probability $p(H)$ for quasi-elastic up-scattering by $\Delta E \geq mg(H_c - H)$, per one UCN collision with the sample-covered part of the trap surface.

Figure 5 shows $p(H)$ for LTF1 at various temperatures. At $T = -90$ °C, p is much lower than for r.t. Fomblin oil, which is also included for comparison. It is also lower than for Fomblin grease, which is about one third of Fomblin oil. The inset shows that these qualitative results also hold for LTF2 at $H = 75$ cm, indicating that for a temperature $T = -90$ °C the new oils are superior to r.t. Fomblin oil in terms of quasi-elastic scattering losses. The temperature dependence shown in the inset is similar to curves for $\nu^{-1}(T)$ where ν is the kinematic viscosity measured in separate experiments. Such a behavior has also been reported for Fomblin oil [19], suggesting that the quasi-elastic scattering observed in these visco-elastic liquids is due to surface modes.

For all materials and temperatures investigated, a quasi-elastic half-width of $\sim$ 20 neV was derived from the slope of the $p(H)$ curves. For Fomblin grease we can compare the magnitude of the mean probability of *up*-scattering, $p_{up} \approx 3 \times 10^{-6}$ for $\Delta E \to 0$, with the probability $p_{down} \approx 1.4 \times 10^{-6}$ for quasi-elastic *down*-scattering by > 4 neV derived from the "long-tail" data. These estimates are consistent within the estimated factor 2 uncertainties, indicating that there is about as much quasi-elastic *up*-scattering as *down*-scattering.

4 Comparison with models

The "long-tail" data (Sect. 2) can be analyzed using a model where quasi-elastic UCN scattering is caused by hydrogen atoms diffusing within an impurity surface layer. If the model applies it should also account for the storage lifetime data since hydrogen is likely to be the dominant source of wall reflection losses as well.

To estimate the surface density n of H-atoms we calculate the reflectivity for a barrier with a thin hydrogenous film on top, neglecting the real part of the layer potential and expanding in terms of the small imaginary potential, using a loss cross section $\sigma_{loss} = 7.4$ barn $(2200/v)$ m/s per H-atom [20] and a density $n_H = 6.7 \times 10^{22}$ cm^{-3} as for liquid H_2O [15]. Averaging over incident angles and the height distribution of UCN density in the gravitational field gives, for a thin film

$$\tau_s^{-1} \approx \tau_n^{-1} + (4g/3v_c^2)(nv\sigma_{loss})(1 + \alpha n + \alpha^2 n^2/3) \times \int (z_0 - z)^{3/2} \mathrm{d}S / \int (z_0 - z)^{1/2} \mathrm{d}V, \quad (1)$$

where $g = 9.81$ m/s^2, $\tau_s \approx 140$ s is the measured storage lifetime, $v_c = 4.51$ m/s the critical velocity for Fomblin grease, and $\alpha = mv_c/\hbar n_H = 1.07 \times 10^{-17}$ cm^2. All UCN have essentially the same small energy, $mgz_0 \approx 17$ neV, given by the fence height $z_0 = 17$ cm. The integrals over (geometric) surface S and volume V of the trap take into account the height dependence of UCN current and density. The solution to (1) is $n = 8.1 \times 10^{16}$ cm^{-2}, which corresponds to a substantial impurity film thickness of ~ 12 nm on top of the grease. The thickness value is reduced if the roughness of the grease substrate is taken into account, and the same applies to p, n and μ (below). Similarly large quantities of hydrogen have also been reported by Arzumanov *et al.* [21] for various solid wall materials.

Using the flat-surface parameters the mean UCN loss probability per reflection becomes $\mu = 5.9 \times 10^{-4}$ and the experimental ratio between p_{down} and μ is

$$r_{exp} = p_{down}/\mu \approx 1.4 \times 10^{-6}/5.9 \times 10^{-4} \approx 0.0024$$

(independently of roughness).

This value will now be compared to an estimate for our model. The classical quasi-elastic incoherent double differential cross section for incident wave number k_0 is [22]

$$\mathrm{d}^2\sigma_{qel}/\mathrm{d}\Omega\mathrm{d}\omega = (\sigma_{inc}k/4\pi^2 k_0)[q^2 D/(\omega^2 + q^4 D^2)], \quad (2)$$

where the classical diffusion model is valid if the momentum transfer $\hbar q = \hbar|\mathbf{k} - \mathbf{k}_0|$ is much less than the inverse of the mean inter-atomic spacing a. Integrating (2) over all angles Ω to obtain $\mathrm{d}\sigma_{qel}/\mathrm{d}\omega$ [15], and then over final wave numbers from zero to k we get

$$\sigma_{qel}(x) = \sigma_{inc} F(x) \quad (3)$$

where $x = k/k_0$, the incoherent scattering cross section of the H-atom is $\sigma_{inc} \approx 80$ barn, and

$$F(x) = (1/8\pi b)\{(x^2 - 1)\ln[(x+1)^2/(x-1)^2] + 4bx\sin\alpha + (x^2 - \cos 2\alpha)\ln[(x^2+1-2x\cos\alpha)/(x^2+1+2x\cos\alpha)] + \sin 2\alpha\{2\arctan[(x^2-1)/(2x\sin\alpha)] + \pi\}\} \quad (4)$$

with $b = 2mD/\hbar$ and $\alpha = 2\arctan b$. The function $F(x)$ is plotted in Figure 6 for $b = 0.057$.

This value of b is based on the estimate [15] that for a thin hydrogenous film the hydrogen diffusion constant D could be as large as for water at 20 °C, $D = 1.8 \times 10^{-5}$ cm^2/s. For $0 < b \ll 1$ and $x < 2$, $F(x)$ resembles the error function with a half-amplitude position slightly above $x = 1$ and a transition width $\Delta x \approx 2.5b$ (from $F = 0.2$ to 0.8). This is the main range of quasi-elastic scattering. For any $b > 0$, UCN quasi-elastic scattering

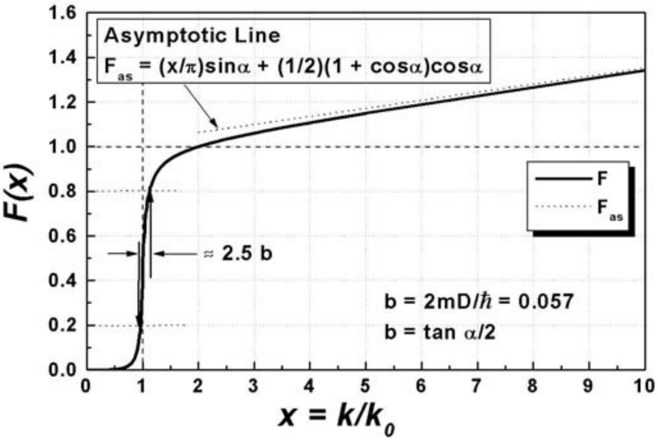

Fig. 6. The quasi-elastic scattering function $F(x)$ (Eq. (4)) and its asymptotic behavior $F_{\mathrm{as}}(x)$ given by (5).

is asymmetric about $\omega = \Delta E/\hbar = 0$, with dominant up-scattering, as reflected in the increase of F above 1.0 for $x > 2$. In the limit $x \gg 1$,

$$F(x) \approx F_{as}(x) = (x/\pi)\sin\alpha + 1/2(1 + \cos\alpha)\cos\alpha \quad [+ \text{ terms of order } x^{-1},\ x^{-3}, \text{ etc.}]. \quad (5)$$

The second term in (5) is a constant close to 1 (for $b \ll 1$) ensuring the unitarity ($\int(\mathrm{d}\sigma_{qel}/\mathrm{d}\omega)\mathrm{d}\omega = \sigma_{inc}$) usually expected for quasi-elastic scattering. However, the first term, which is $\sim b$, breaks unitarity (without violating detailed balance), as a consequence of the kinematical factor k/k_0 in (2). It increases linearly with $x = k/k_0$ until, for $k \sim a^{-1}$, *i.e.* far outside the UCN range, classical diffusion breaks down and the growth of F with k is limited by the Debye-Waller factor for jump diffusion. In the limit $k_0 \to 0$, the integral quasi-elastic scattering of UCN on diffusing atoms shows the same divergence $\sim 1/k_0$ as thermal inelastic scattering and nuclear capture.

For the trap geometry of Figure 1 the long tail is due to UCN in a band Δk from $\sim 0.62k_0$ to $\sim 0.95k_0$ where $k_0 = mv_0/\hbar$ and $v_0 = (2gz_0)^{1/2} = 1.83$ m/s is the UCN velocity at the trap bottom. The lower value $0.62k_0$ is determined by the elevation 6.5 cm of the lowest point of the exit cross section; the upper value $0.95k_0$ is for UCN with efflux time constant $\tau \approx 30$ s, which is just barely compatible with the measured $\tau \approx 40$ s.

From (3) and (4) it follows that the partial scattering cross section with final states in the band Δk is $\sim 16\%$ of σ_{inc}, *i.e.* $\sigma_{qel} \approx 13$ barn for $\sigma_{inc} \approx 80$ barn. Comparing $\Delta\sigma_{qel}$ with the mean loss cross section per H-atom, $\sigma_{loss} = 1.63 \times 10^4$ barn $\langle 1/v \rangle$ m/s $= 1.07 \times 10^4$ barn, we get

$$r_{\mathrm{model}} = \Delta\sigma_{qel}/\sigma_{loss} \approx 0.0010$$

(independently of roughness).

If the presence of a given amount of hydrogen on the walls is the common origin of the long tail and of the wall losses then we expect that this estimate is compatible with the experimental ratio $r_{exp} \approx 0.0024$ obtained above.

Thus, the model of hydrogen diffusion within a hydrogenous impurity layer on the Fomblin grease surface is consistent with the data within a factor 2.5.

For the liquid wall materials, we can compare the data to the analysis [16] of UCN quasi-elastic scattering on visco-elastic surface waves. The calculated distribution is non-Lorentzian and strongly peaked at energy transfers $\Delta E < 1$ neV, but has an up-scattering tail reaching into the range > 10 neV with a slow decrease. These higher energy transfers are selectively reflected in the data due to the high-energy bias of Al foil transmission and can explain the measured half-width of ~ 20 neV. Measurements showing directly that transmission through foils sets in very slowly above the critical edge have been reported by Serebrov *et al.* in [19].

5 Summary and conclusions

In storage experiments with Fomblin grease coated UCN traps at room temperature we observed both sides of quasi-elastic energy transfer: down-scattering and up-scattering. To explain the measured slow UCN efflux from trap 1 we obtained a mean probability $p_{down} \approx 1.4 \times 10^{-6}$ for down-scattering by ≥ 4 neV in a wall collision. Within the uncertainty of estimating p_{down} and p_{up} from the data this value is consistent with the up-scattering probability $p_{up} \approx 3 \times 10^{-6}$ determined in a different trap (2) for a different sample of Fomblin grease.

To interpret the magnitude of p_{down} we used the model [15] of quasi-elastic UCN scattering on hydrogen diffusing in an impurity layer on the surface. If the model applies the amount of hydrogen needed to explain the measured storage lifetime in trap 1 should also account for the integral "long-tail" intensity. The agreement within a factor 2.5 is satisfactory.

From the energy transfer (ΔE) dependence of quasi-elastic scattering on Fomblin grease we estimated a value of ~ 20 neV for a decrease of integral up-scattering by a factor 2. This value is compatible with the high-energy distribution for the hydrogen diffusion model [15]. About the same slope was obtained for the liquid wall materials investigated in trap 2: Fomblin oil at 25 °C and two kinds of "low temperature Fomblin" (LTF) in the range -110 to $+25$ °C (Fig. 5), in general agreement with calculations [16] for visco-elastic surface waves at large ΔE.

In conclusion, the data are consistent with the models of hydrogen diffusion (for the grease data) and surface waves (for the liquid wall coatings). No "anomalies" have to be invoked to explain these data, while the scattering at larger ΔE on solid walls, *e.g.* on beryllium, is still under discussion [14,19]. We have also found that the new low-temperature oils show very little quasi-elastic scattering at temperatures < 180 K and are, therefore, good candidates for a new type of UCN trap with significantly lower losses than for the best material known so far: Fomblin oil, which is, however, too viscous to be used below $T \approx 4$ °C [7]. If further tests will be positive, an improved measurement of the neutron lifetime appears possible.

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