Measurements of ultracold neutron lifetimes in solid deuterium

C. L. Morris, J. M. Anaya, T. J. Bowles, B. W. Filippone, P. Geltenbort, R. E. Hill, M. Hino, S. Hoedl, G. E. Hogan, T. M. Ito, T. Kawai, K. Kirch, S. K. Lamoreaux, C.-Y. Liu, M. Makela, L. J. Marek, J. W. Martin, R. N. Mortensen, A. Pichlmaier, A. Saunders, S. J. Seestrom, D. Smith, W. Teasdale, B. Tipton, M. Utsuro, R. A. Young, and J. Yuan

Los Alamos National Laboratory, Los Alamos, NM 87544, USA
Kellogg Radiation Laboratory, California Institute of Technology, Pasadena, Ca 91125, USA
Institut Laue-Langevin, BP 156, F-3804 Grenoble cedex 9, France
University of Kyoto, Kyoto 606-8501, Japan
Princeton University, Princeton, NJ 08544, USA
Virginia Polytechnical Institute and State University, Blacksburg, Va 24061, USA
North Carolina State University, Raleigh, NC 27695, USA
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We present the first measurements of the survival time of ultracold neutrons (UCNs) in solid deuterium (SD$_2$). This critical parameter provides a fundamental limitation to the effectiveness of superthermal UCN sources that utilize solid ortho-deuterium as the source material. Superthermal UCN sources offer orders of magnitude improvement in the available densities of UCNs, and are of great importance to fundamental particle-physics experiments such as searches for a static electric dipole moment and lifetime measurements of the free neutron. These measurements are performed utilizing a SD$_2$ source coupled to a spallation source of neutrons, providing a demonstration of UCN production in this geometry and permitting systematic studies of the influence of thermal up-scatter and contamination with para-deuterium on the UCN survival time.

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Neutrons with kinetic energies less than 340 neV (corresponding to a temperature T<5 mK) can be trapped in material bottles and are referred to as ultracold neutrons (UCNs). UCN densities at reactor sources have gradually increased with reactor power and improved techniques for extracting the UCN flux. The highest bottlenecked densities reported in the literature, 41/cm$^3$, have been obtained at the Institut Laue-Langevin (ILL) reactor in Grenoble [1].

Measurements of the neutron electric dipole moment [2,3] and the neutron lifetime [4-6] attest to the utility of bottled UCNs for fundamental experiments with neutrons. UCNs may prove useful in improved measurements of angular correlations in neutron beta-decay [7,8], although experiments of this kind utilizing UCNs have not yet been performed. All of these experimental programs have been limited by the available densities of UCNs.

Superthermal UCN production, where the production rate of UCNs due to down-scattering in energy is larger than the combined up-scatter and nuclear-absorption rates in the material, was first proposed in 1975 by Golub and Pendlebury [9] in superfluid $^4$He and experimentally investigated shortly thereafter [10,11]. In this process phonon creation in the liquid is used to down-scatter cold neutrons to the UCN regime, while up-scattering is suppressed by maintaining the superfluid at sufficiently low temperature. Because $^4$He has no nuclear absorption, the only limitations to the density of UCNs accumulated are wall losses and neutron beta decay. The production of UCNs by this process has been observed and agrees with theoretical expectations [12,13,14,15,16,17,18].

While superfluid $^4$He is an excellent superthermal converter, a few other materials, such as solid deuterium (SD$_2$), satisfy the criteria for superthermal production. The limiting UCN density, $\rho_{UCN}$, one can obtain using a SD$_2$ source is given by the product of the rate of UCNs production in the solid, $R$, and the lifetime of UCNs in the solid, $\tau_{SD}$: $\rho_{UCN} = R\tau_{SD}$. A storage bottle opened to such a source will come into density equilibrium with the density in the solid. This led to the proposal of a thin-film source where the inside of a neutron bottle is coated with a thin layer of SD$_2$ and the bottle is embedded in a cold neutron flux [21]. The volume comes into equilibrium with the UCN density with a time constant for the coupled system, $\tau$, given by,

$$\tau = \frac{\tau_{SD} V}{V_{SD}},$$

where $V_{SD}$ is the volume of SD$_2$ in the source and $V$ is the total volume of the storage bottle. A valuable summary of SD$_2$ thin-film sources is presented in [21]. The effects of gravity and of the potential of the solid as well as UCN losses other than absorption in the film have been neglected in this expression but do not fundamentally alter the above picture. Ultimately, the limit to the UCN density is established by the trade-off between the cold-neutron flux intensity and energy distribution (which determine the production rate) and the heat deposited by neutrons and gamma rays in the SD$_2$ and bottle walls, because $\tau_{SD}$ is a strong function of temperature. However,
the predicted production rates in SD$_2$ [20] and lifetimes have not yet been quantitatively verified. Efforts to utilize SD$_2$ sources at reactors identified possible gains but suffered from problems with cooling the solid at full reactor power [13].

Pokotilovski pointed out the advantages of UCN production in SD$_2$ at pulsed neutron sources [23] and showed the UCN densities 4–5 orders of magnitude greater than existing reactor-based UCN sources might be possible. More recently, the use of spallation as a pulsed source has been suggested [24,25]. In a spallation UCN source, cold neutrons are produced by moderating spallation neutrons produced in a heavy target by a medium-energy pulsed proton beam. These cold neutrons are used to drive a SD$_2$ superthermal converter. In a spallation target, the amount of heating for each neutron is lower than in a reactor, allowing higher neutron densities in the vicinity of a spallation target to be achieved. Even higher neutron densities can be obtained by pulsing the proton beam and valving off the UCN storage volume from the production volume when the beam is off and by using the time when the beam is off to remove heat from the deuterium. In this case, the maximum UCN density that is produced is limited only by the impulse heating of the SD$_2$. Experiments with the stored UCNs can be performed while the beam is off, eliminating the backgrounds due to capture gammas found near continuously operated reactor sources.

In order to test the concept of a SD$_2$-based spallation UCN source, we have built a test source that we have operated with single pulses of protons produced by the LANSCE 800 MeV proton accelerator at Los Alamos National Laboratory. In this letter we report the first measurements of $\tau_{SD}$, the lifetime of UCNs in SD$_2$. Our measurements clearly demonstrate the critical influences of heating and para-deuterium contamination on the UCN lifetime, and provide a quantitative foundation for the development of SD$_2$ superthermal sources.

A schematic view of our apparatus is shown in Fig. 1. Spallation neutrons were produced in a tungsten target with short (typically less than 160 ns long) pulses of 800 MeV protons from the LANSCE accelerator. The fast neutron flux was contained and amplified using (n,2n) reactions in a layer of beryllium surrounding the spallation target. The spallation neutrons were moderated and cooled in a thin layer of polyethylene surrounding a $^{58}$Ni-coated stainless steel guide tube with an inner diameter of 7.8 cm. The polyethylene and the bottom of the guide were cooled with liquid helium, and a layer of deuterium was frozen on the inside of the guide. UCNs produced in the SD$_2$ were confined by the guide tube and could be directed through a series of valves to the UCN detector. Neutrons were detected in a 5-cm-thick multiwire chamber detector filled with a mixture of $^3$He at 5 mbar and CF$_4$ at 1 bar. The low $^3$He pressure and the large bend angle in the guide resulted in a high degree of selectivity for detecting UCNs in the apparatus. Data were acquired using a multi-scalar that was started by the proton beam passing through a toroidal pick-up coil and that scaled the count rate from the UCN detector.

Several effects limit the lifetime of UCNs in SD$_2$: up-scatter from phonons in the solid [20], up-scatter from para-deuterium molecules in the solid [22], absorption on deuterium, and absorption on hydrogen impurities in the solid. Model calculations exist for the contribution of all of these effects on the UCN lifetime in the solid. The total loss rate is a sum of contributions from each of the sources listed above:

$$\frac{1}{\tau_{SD}} = \frac{1}{\tau_{phonon}} + \frac{1}{\tau_{para}} + \frac{1}{\tau_{Dabs}} + \frac{1}{\tau_{Habs}}, \quad (2)$$

with the loss rate due to phonon up-scatter having different contributions from the ortho- and para-deuterium in the solid. Establishing the experimental basis to validate these models as both accurate and complete is essential for the design of a UCN source based on the superthermal production mechanism in solid ortho-deuterium.

SD$_2$ was frozen in the lower part of the cryostat using a helium transfer refrigerator. The temperatures of the lower guide walls and of the liquid-helium cryostat were monitored with an array of silicon diodes mounted on the guide and the aluminum cryostat walls. The measured temperatures tracked the vapor pressure curve of SD$_2$ well at higher temperatures. The temperature of the solid at lower temperatures was obtained by averaging the temperatures of two diodes mounted on the outside of the guide wall. Later measurements made with diodes embedded in the solid indicate these measurements are accurate to 1 K.

Both the hydrogen contamination and the para-
fraction in the SD$_2$ were measured by means of rotational Raman spectroscopy on a gaseous sample taken by warming the deuterium after the measurement [22]. These measurements yielded values for the HD concentrations in the gas that varied from 0.2–0.3% with an uncertainty of about 0.1%. Other contamination was removed by a palladium membrane in the D$_2$ gas system, before introducing D$_2$ to the cryostat. The para-fraction was controlled by converting the D$_2$ to a near thermal equilibrium otho/para ratio in an iron-hydroxide-filled cell [22] cooled to a temperature at or slightly below the triple point. In this way the para-fraction was reduced from room-temperature equilibrium value of 33% to 2–4%. Intermediate values were obtained by mixing deuterium at room temperature equilibrium with converted deuterium before freezing. The precision of the para-fraction measurements was typically of order of 1%. The solid volume was measured by integrating the flow of gas while growing the solid. The volume was checked when the solid was warmed and the gas was returned to a buffer volume. The uncertainties in the pressure dependence of the calibration of the flow meter (20%), and uncertainties in the guide volume and temperature combined to lead to an uncertainty in the solid volume of about 20%.

The sensitivity of the apparatus to UCNs was demonstrated by measuring neutron arrival times with and without a $^{58}$Ni-coated 0.024-cm-thick aluminum foil in place at location C (in Fig. 1). These data are shown in Fig. 2. The number of counts, arriving in a time window between 0.5 sec and 10 sec, with the foil opened was 3% of the number with the foil opened. About half of these could be attributed to UCN leakage through the gap around the outside of the valve, and the rest were neutrons with normal velocities sufficient to penetrate the potential barrier provided by the $^{58}$Ni. These data unambiguously demonstrate that the signal in the $^3$He detector at the end of the guide was predominantly due to UCNs.

If gravity, wall losses, the SD$_2$ potential, and transport effects are neglected, and if the SD$_2$ is thin enough so that its volume is uniformly sampled by the neutrons, the lifetime of neutrons stored in a bottle in contact with SD$_2$ is given by equation (1). We have used this idea to measure the UCN lifetime in SD$_2$. As depicted in Fig. 1, valves B and C were open for these measurements. UCNs were stored in the bottle between the end of the guide and valve A, which includes the SD$_2$. The number of neutrons detected in a 10 sec wide time gate after the time, $t$, when valve A was opened was measured, typically for $t=0.5, 1, 2, 4$ seconds. These data were fitted with the form $c_o e^{-t/\tau}$. The parameters $c_o$ and $\tau$ were varied to produce the best fit for various conditions of the SD$_2$. Background was subtracted by a fitting linear function to the data between 20 sec and 65 sec after the trigger and extrapolating under the data. The $t_{SD}$ were extracted from the $\tau$ using a lookup table of $t_{SD}$ vs $\tau$ generated by fitting lifetime scans modeled with Monte Carlo for different $t_{SD}$ that were analyzed in the same fashion as the data.

In the Monte Carlo transport, the full experimental geometry, gravitation, the SD$_2$ potential (108 neV), and wall collisions in the guide tube have been taken into account. There are a number of parameters that must be determined from the experimental data in order to extract $t_{SD}$: the probability of UCN loss and nonspecular reflection for each collision with the guide wall, the SD$_2$ elastic scattering length, and the physical configuration of the SD$_2$ frozen on the walls (SD$_2$ can freeze as a flat “pancake” on the bottom of the guide, or coat the entire He-cooled surface at the bottom of the guide, in a “bucket”-shaped configuration).

Because we used very thin samples of SD$_2$ for most of the lifetime measurements (0.4 cm), we were quite insensitive to the scattering length for UCNs in SD$_2$. The saturation of UCN production we observe for thicker samples (up to ~6 cm of sample thickness) is consistent with the large scattering length calculated by Hill et al. [27]. However, our extracted lifetime results change by less than 10% for scattering lengths as small as 0.5 cm. We therefore used the theoretical value of 8 cm for the elastic scattering length for all of the results presented here.

The ratio of diffuse to specular reflections for wall collisions was adjusted to fit time-of-arrival spectra (see Fig. 2). The fraction of wall losses per wall collision and the shape of the SD$_2$ ice were adjusted in a combined fit to reproduce the volume dependence for all of the low-temperature data. The extracted values for the guide parameters (0.025 for the ratio of diffuse to specular reflections) and wall losses ($2.0\times10^{-3}$) were consistent with those extracted from an independent set of measurements of guide transmissions and holding times at
the ILL. The lifetimes due to absorption on deuterium and hydrogen were calculated using the known amount of hydrogen contamination, the tabulated thermal cross sections and the $1/v$ dependence of the cross sections. In addition the lifetimes in solid ortho- and para-deuterium due to temperature up-scattering were taken from the literature. The lifetime in solid para-deuterium due to molecular transitions was treated as a free parameter and found to be $\tau_{\text{SD, para}} = 1.2 \pm 0.2$ ms, consistent with a calculation that gives $\tau_{\text{SD, para}} = 1.5$ ms.

Results for UCN lifetimes $\tau_{\text{SD}}$ in SD$_2$ as a function of the SD$_2$ temperature and para/ortho-fractions are shown in Fig. 3. The difference between the solid and dashed lines demonstrates the the need to include the effect of deuterium vapor in the guide on the lifetime at higher temperatures. With this correction, the measured lifetimes agree well with theoretical predictions of the up-scatter rate. The main contributions to the UCN lifetime in SD$_2$ have been measured and are quantitatively understood. These data demonstrate that UCN sources based on SD$_2$ converters can provide the promised large gains over existing sources.

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* Present address:Research Center for Nuclear Physics, Osaka University, Japan
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