Calculation of the ultracold neutron upscattering rate from para-deuterium bound in a lattice

C.-Y. Liu¹, A.R. Young¹, and S. K. Lamoreaux²
1. Princeton University, Physics Dept., Princeton, N.J.
2. University of California, Los Alamos National Laboratory, Physics Division P-23, Los Alamos, NM 87545
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The ultracold neutron (UCN) upscattering rate from para-deuterium contaminated solid D₂ near T = 0 K is calculated and found to be proportional to the atomic fraction of para contaminant (x_{para}) as \( \tau_{up}^{-1} \approx 700 x_{para} \sec^{-1} \). With \( x_{para} = 0.33 \), corresponding to room temperature equilibrium, the upscatter time is 4.4 msec. The implication is that the para-deuterium contaminant is the temperature-independent energy source responsible for the observed upscattering rate in the Los Alamos spallation driven UCN source.

I. INTRODUCTION

The possibility to use low neutron absorption cold materials, such as solid deuterium, as a moderated source of ultracold neutrons (UCN) has been long discussed [1]. However, as pointed out by Golub and Pendlebury [2], one can in fact attain a much higher UCN density than implied by the thermal equilibrium arguments given in [1]: if we consider UCN produced by inelastic downscattering (in energy) of cold or thermal neutrons, the reverse process can be relatively suppressed. In the case of moderator with two energy levels, a ground state and an excited state separated by an energy \( \Delta \), the upscatter rate of a UCN with energy \( E_{ucn} \) and the downscatter rates of a neutron with energy \( E_{ucn} + \Delta \) rates are related according to microscopic reversibility:

\[
\sigma_{up} = \sigma_{down} e^{-\Delta / kT}. \tag{1}
\]

The implication of this result is that, at very low temperatures, UCN can be accumulated in a material until the rate of production \( P \) equals the rate of loss, \( 1/\tau \), due to nuclear absorption or leakage from a storage volume that surrounds the converter material; the density in equilibrium is

\[
\rho = P \tau \tag{2}
\]

This is the so-called “superthermal” UCN production process, first described by Golub and Pendlebury in relation to their proposed superfluid \(^4\)He UCN source. [2,3] The theory of the superthermal process for superfluid \(^4\)He has been now very well established, both by observation of upscattered UCN from a storage volume [4], and by direct measurement of the UCN density in a nearly losses superfluid \(^4\)He filled magnetic trap [5].

The possibility of the use of other materials of low neutron absorption cross section as a superthermal source was first investigated by Golub et al. [6] after Mr. Kilvington observed the production of UCN in a prototype superfluid \(^4\)He source, operated at the Institut Laue-Langevin, even when it was empty of \(^4\)He; the explanation by Golub was that there was a film of dirt on the inner surface of the UCN converter/storage vessel and this dirt was operating as a superthermal source. This led to the proposal of the “thin film superthermal source”: Unlike the case of \(^4\)He, all other materials have some neutron absorption cross section. For materials like solid D₂, the absorption lifetime is 120 ms, compared to the effective lifetime of about 880 sec of a UCN in superfluid \(^4\)He at low temperature, in which case \( \beta \)-decay is the ultimate limiting loss mechanism. The idea of the thin film source is that the UCN density in a storage volume of volume \( V \) that contains some converter material is independent of the volume of the converter material \( V_c \) because the total production rate is proportional to \( V_c \), while the net effective lifetime \( \tau \) is proportional to \( 1/V_c \) (in the limit where upscattering and absorption in the converter material dominates all other loss mechanisms), and \( V_c \) cancels in Eq. (2), \( V \) determines the time constant for filling the volume \( V \) to an equilibrium density.

The figure of merit for a superthermal source is given by \( P \tau \) for the converter material. Superfluid \(^4\)He is at least an order of magnitude better than any other material; in this case, even though \( P \) is relatively low, the long intrinsic lifetime more than makes up the difference in production rate compared to other materials. However, there are situations where use of a less efficient material is warranted. For example, a thin film source might be better suited near the core of a nuclear reactor because less converter material is required to achieve the intrinsic maximum UCN density.

Another situation where a material with a large \( P \) but short \( \tau \) was first pointed out by Pokotilovski [6] in connection with pulsed neutron sources, such as spallation sources. The idea is that the converter material is connected to a UCN storage volume only for a short time during and after the neutron pulse, after which a valve is closed, separating the conversion and storage regions. In this case, one can enjoy a relatively large \( P \) together
with an net effectively large $\tau$. Serebrov elaborated upon this idea with the proposal of a “UCN Factory”: following this lead, we developed a prototype UCN source that turns out to follow the original Pokotilovski idea more closely, but ours incorporates a cold neutron flux trap, and vertical UCN extraction from the solid D$_2$ converter to counteract the solid deuterium positive potential step.

In our recent studies of solid deuterium as a spallation driven generator of ultracold neutrons, the apparent production rate is roughly an order of magnitude less than expected, and there is a lack of temperature dependence on the net production rate, contrary to the prediction by Golub et al. [13] The reason for the reduction appears primarily due to a loss of UCN before they can exit the solid deuterium. By elimination of various loss sources within the material that can account for the temperature independent loss, we came to the conclusion that the UCN loss is likely due to upscattering from para-deuterium contaminant with solid (ortho-) deuterium converter material; in addition, anecdotal evidence for this was offered by Serebrov. The following calculation estimates the upscatter rate and was motivated for incorporating a para/ortho converter [3]. The preliminary results indicate the following calculation is correct; the data and analysis will be presented in a full publication by the collaboration. We felt it important, however, to make available our initial estimate of the upscatter rate due to the para contaminant.

II. PROPERTIES OF D$_2$ AND ASSUMPTIONS IN THE CALCULATION

The deuterium molecule exists in the ground state as a total nuclear spin $S = 0$ and rotational angular momentum $J = 0$, and is referred to as ortho–deuterium. The first excited state has $S = 1$ and $J = 1$, and the energy relative to the ground state is

$$E_0 = 7 \text{ meV}. \quad (3)$$

We refer to this state as para–deuterium.

Of course, at high temperature, the $S,J$ states are occupied according to their degeneracy and Boltzmann factor; just considering the nuclear states, the ortho-states ($S = 0, 2$) and para-states ($S = 1$) are occupied at high temperatures according to their spin degeneracies:

$$x_{\text{para}} = \frac{3}{1 + 3 + 5} = 0.33; \quad x_{\text{ortho}} = \frac{1 + 5}{1 + 3 + 5} = 0.67 \quad (4)$$

because the degeneracy is given by $2S + 1$. The rotational states are coupled strongly to the lattice and we can assume the $J$ populations follow the Boltzmann distribution with rapid equilibration. On the other hand, the transitions between ortho and para is very slow under normal conditions, and we can assume that the room temperature composition persists as the D$_2$ sample is cooled and solidified, and is rather permanent in our experiment.

We can assume the sample is at absolute zero; we therefore can neglect upscattering from the vibrations of the lattice which represents a different upscatter mechanism, roughly independent of upscattering from the para contaminant. Furthermore, we can make an approximation that the molecules are fixed in the lattice and neglect recoil effects; this approximation is valid because a) $E_0 < \theta_D$, the Debye temperature, b) the momentum transfer is in the form of angular momentum from the molecular rotation to the outgoing wave, c) the UCN energy $E_i << \theta_D$, making direct excitations of the lattice unlikely. Finally, we assume that all the para molecules are in the $J = 1$ rotational state, while the ortho molecules are in the $J = 0$ state. This final assumption is valid because the rotational levels, unlike the nuclear spin state, are closely coupled to the thermal bath, as mentioned above.

Normally, we assume that $P$–wave scattering is suppressed for cold and ultracold neutrons. However, in this case, the scattering is from a molecule; the separation between the deuterium atoms in the molecule is comparable to the wavelength of an outgoing upscattered UCN (with final energy 7 meV) and we do not have the usual $P$–wave scattering suppression found in nuclear scattering from a single isolated nucleus.

III. UPSCATTER RATE

Much of Young and Koppel (YK) [13] is irrelevant to this calculation and in addition is not entirely transparent. Our problem is simple enough to begin from first, very basic, principles. We will see than only a single matrix element from YK is required for our calculation, as subject to the above constraints and approximations.

Starting with Fermi’s Golden Rule for transitions to a continuous state (see [10], Sec. 43)

$$dw_{fi} = \frac{2\pi}{\hbar} \left| U_{fi} \right|^2 \delta(E_f - E_i) d\nu_f \quad (5)$$

where $dw_{fi}$ is the transition rate between the levels $f,i$, $d\nu_f$ is the density of final states, and $U_{fi}$ is the matrix element of the Hamiltonian between the initial and final states,

$$U(\vec{r}) = -\frac{2\pi \hbar^2}{m} \left( a_1 \delta(\vec{r} - \vec{r}_1) + a_2 \delta(\vec{r} - \vec{r}_2) \right) \quad (6)$$

where 1, 2 label the two atoms of the molecule, $m$ is the neutron mass, and $a_{1,2}$ are the scattering lengths, chosen to represent the ortho- or para- states as required.

First, let us take the incoming and outgoing neutron states as plane waves (as did YK). The incident (UCN)
wave must represent the UCN density $\rho_n$ in a normalization volume $V$. Thus,
\begin{equation}
\psi_{i,n} = \frac{1}{\sqrt{V}} \sqrt{\rho_n} e^{i\vec{k}_f \cdot \vec{r}} = \sqrt{\rho} \psi_i \tag{7}
\end{equation}

where the factor $1/\sqrt{V}$ normalizes the incident wave to the volume $V$, the second factor gives the total UCN number $n = \rho_n V$ within $V$, the subscript $i,n$ means incident wave, normalized, and $\psi_i$ conforms to the states used by YK.

The outgoing wave can be written
\begin{equation}
\psi_{f,n} = \frac{1}{\sqrt{V}} e^{-i\vec{k}_f \cdot \vec{r}} = \psi_f/\sqrt{V}. \tag{8}
\end{equation}

The density of final states $\vec{k}_f$ is
\begin{equation}
d\nu_f = \frac{V d\vec{k}_f}{(2\pi)^3} = \frac{Vk^2_d dk_f d\Omega}{(2\pi)^3}. \tag{9}
\end{equation}

In Fermi’s Golden rule, the $\delta$-function ensures energy conservation. In the present case,
\begin{equation}
E_f - E_i = h^2 k_i^2 /2m - h^2 k^2_f /2m - E_0 \tag{10}
\end{equation}

and we have $\delta(f(k_f))$. Recall the well-known relationship,
\begin{equation}
\delta(f(x)) = \frac{1}{|f'(x_0)|} \delta(x - x_0) \tag{11}
\end{equation}

where $x_0$ is the zero of $f(x)$. A further assumption $\vec{k}_i = E_i = 0$ is valid for UCN in this case. Then the momentum transfer $\vec{k} = \vec{k}_f$, in YK’s notation.

Therefore, the upscatter rate is
\begin{equation}
dw_{fi} = \frac{2\pi}{\hbar} (\frac{2\pi \hbar^2}{m})^2 
\times | \langle \psi_f, J' = 0, S' = 1 \rangle |^2 \frac{m}{\hbar^2 k_f} 
\times \delta(k_f - \sqrt{2mE_0/\hbar^2}) \frac{V k^2_d dk_f}{(2\pi)^3} \tag{12}
\end{equation}

which must be integrated over $k_f$; this is easily done, and we write the matrix element $\sum |a_i|^2$, yielding a total rate, taking $V x_{para} \rho_{D_2}$ scattering molecules in $V$,
\begin{equation}
w_{fi,tot} = 4\pi V x_{para} \rho_{D_2} \rho_{ucn} \frac{\hbar}{m} k_0 | \sum a_i |^2 \tag{13}
\end{equation}

where
\begin{equation}
k_0 = \sqrt{2mE_0 \hbar^2} = 1.83 \times 10^6 \text{cm}^{-1} \tag{14}
\end{equation}

and
\begin{equation}
\frac{\hbar}{m} k_0 = v_0 = 1.2 \times 10^5 \text{cm/s}. \tag{15}
\end{equation}

We further note that $w_{fi,tot}/V = \dot{\rho}_{ucn}$ so we have a loss rate of
\begin{equation}
\Gamma = 4\pi x_{para} \rho_{D_2} v_0 | \sum a_i |^2 \tag{16}
\end{equation}

Thankfully, YK have calculated the matrix elements. We only need
\begin{equation}
| \langle k_f, J' = 0, S' = 1 \rangle |^2 = 3 \sum a_{inc}^2 \frac{C^2(1011 : 00)}{A_0} \tag{17}
\end{equation}

from Eq. (A5) of KY and Appendix B. Also, $(a = .742 \text{Å} \text{ cm})$ so $k_0a/2 = 0.68$

\begin{equation}
|A_{01}|^2 = |2ij_1(k_0a/2)|^2 = 4 \times j_1(k_0a/2)^2 = 4 \times (.21)^2 = .18 \tag{18}
\end{equation}

so
\begin{equation}
| \sum a_i |^2 = 0.13a_{inc}^2. \tag{19}
\end{equation}

Taking $a_{inc} = 4.04 \times 10^{-13} \text{ cm}$ gives
\begin{equation}
\Gamma = 3.2 \times 10^{-20} x_{para} \rho_{D_2} \text{ cm}^{-1} \tag{20}
\end{equation}

and, with $x_{para} = 0.33$ corresponding to room temperature equilibrium, and $\rho_{D_2} = 3 \times 10^{22} / \text{cm}^3$ gives
\begin{equation}
\Gamma = 316 \text{ sec}^{-1}; \quad \tau = 3.2 \text{ ms}. \tag{21}
\end{equation}

Finally, zero-point motion of the lattice leads to a smearing-out of the final state, and a reduction in the cross section. This effect is given by the Debye-Waller factor, which for solid deuterium is a factor of 0.72 for a momentum transfer corresponding to 7 meV. [5] The net upscatter rate and lifetime is thus
\begin{equation}
\Gamma = 221 \text{ sec}^{-1}; \quad \tau = 4.4 \text{ msc}. \tag{22}
\end{equation}

IV. DISCUSSION

The important implication of this calculation is that the upscatter rate from ortho-deuterium is independent of temperature and persists at absolute zero. The agreement between this simplified calculation and experiment is satisfactory and appears to have identified a principal limitation to the present Los Alamos UCN source density.

The surprising result is that the $P$–wave scattering is significant: the outgoing plane wave state carries away the rotational angular momentum of the molecule. This is possible because the impact parameter, corresponding to the molecular size, is comparable to the outgoing neutron wavelength, as is illustrated in Eq. [5].
Contamination of a solid deuterium UCN source by the para-molecular state will severely limit the attainable UCN density. Given the stability of the para-deuterium state (0.02%/hr conversion rate in the solid) it is clear that the gas must be converted before freezing. The details of the Los Alamos converter will be published, but it follows standard techniques. Our preliminary results support the above calculated rate.

Our result has serious implication for solid deuterium sources operated close to the core of a nuclear reactor. In this situation, the gamma flux will be sufficient to drive molecular excitations, and the ortho-para ratio will likely be near the high temperature equilibrium. This implies that solid deuterium might be useless in such an environment, although the presence of free charge in the lattice might catalyze the conversion. We are presently investigating these and other effects expected in a high radiation environment. We further point out that the radiation levels in our proposed spallation driven source are many orders of magnitude less than a reactor so we expect no problems in this regard.

A more complete calculation of the upscattering rate and related issues is in progress by the authors.

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