UCN upscattering rates in a molecular deuterium crystal

C.-Y. Lin\textsuperscript{1}, A.R. Young\textsuperscript{1}, and S. K. Lamoreaux\textsuperscript{2}

\textsuperscript{1.} Princeton University, Physics Dept., Princeton, NJ 08544
\textsuperscript{2.} University of California, Los Alamos National Laboratory, Physics Division P-23, Los Alamos, NM 87545

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A calculation of ultra-cold neutron (UCN) upscattering rates in molecular deuterium solids has been carried out, taking into account intra-molecular excitations and phonons. The different molecular species ortho-D\textsubscript{2} (with even rotational quantum number J) and para-D\textsubscript{2} (with odd J) exhibit significantly different UCN-phonon annihilation cross-sections. Para- to ortho-D\textsubscript{2} conversion, furthermore, couples UCN to an energy bath of excited rotational states without mediating phonons. This anomalous upscattering mechanism restricts the UCN lifetime to 4.6 msec in a normal-D\textsubscript{2} solid with 33% para content.

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The low density of ultra-cold neutrons (UCN) available using conventional cold moderators at nuclear reactors has long been the main constraint in the pursuit of high precision measurements of neutron $\beta$-decay with UCN [1]. To pursue the possibility of utilizing this simple hadronic system for tests of weak interaction theories, Golub and Pendlebury [2] proposed a way to increase UCN production through the exchange of energy between a cold neutron bath and the phonons in certain cold moderators such as super-fluid $^4$He, with large neutron scattering cross-sections and small neutron absorption cross-sections. Superthermal UCN sources exploit the characteristics of low temperature substances, in which a large number of phonon modes are available for neutron downscattering, while the number of phonons present which can upscatter UCN is suppressed. Ideally, the density of UCN produced in such a source is limited either by nuclear absorption in the moderator as is the case for solid deuterium, or the neutron lifetime itself as is the case for superfluid $^4$He.

A solid D\textsubscript{2} UCN superthermal source [3] is under development at the Los Alamos Neutron Science Center. Preliminary estimates [4] promise gains in the available UCN density as high as two orders of magnitude over existing UCN facilities. To characterize its performance, calculations of essential physical parameters, such as scattering cross-sections, UCN residence time, etc., with more detailed and accurate models than those presently available are in increasing demand. In this report, we demonstrate that in the presence of even very small concentrations of para-D\textsubscript{2} (with total nuclear spin 1) can dominate the UCN upscattering rate, overwhelming the usual phonon annihilation mechanism. This results in greatly reduced UCN lifetimes in the solid and orders of magnitude reductions in the achievable UCN density.

The deuterium molecule is a two-body system with the quantum properties of identical bosons. Its nuclear spin wave-function couples to molecular rotational states with same parity to preserve the symmetry of the wave-function under permutation of identical particles. Koppel and Young [5] calculated the neutron scattering cross-section of this molecular system, taking into consideration induced transitions between molecular rotational and vibrational states. In their formulation, an incoherent approximation was used and translational coordinates were assumed to commute with inter-molecular degrees of freedom. The derived double differential cross-section in which the interference term is neglected has the form, for UCN with incident wavenumber $k$ and final wavenumber $k'$,

$$
\frac{d^2\sigma}{d\Omega dk'} = \frac{1}{2\pi\hbar^2} \int_{-\infty}^{\infty} dt \sum_{l} e^{-i\kappa r_l(0)} e^{i\kappa r_l(t)} \langle \phi_{kl} | >_{\text{trans}} \sum_{J} P_{JS} \sum_{J'} S_{JJ'}(2J' + 1)e^{i(E_{J'} - E_{J})t/\hbar} \times \sum_{n=0}^{\infty} \frac{e^{i\omega n\tau}}{n!} \left( \frac{\hbar^2 \kappa^2}{2MD_2}\right)^n \sum_{l=|J'-J|}^{J'+J} |A_{nl}|^2 C^2(JJ';00), \tag{1}
$$

where $\hbar\kappa$ is the momentum transfer of the scattered neutron, $P_{JS}$ is the population of the initial molecular state with a total nuclear spin $S$ and rotational quantum number $J$, $E_J = 7\text{meV} \times J(J+1)/2$ is the rotational spectrum, $\hbar\omega$ is the inter-molecular vibrational energy with $n$ characterizing the number of vibrational energy quanta, and $C(JJ';00)$ is a Clebsh-Gordon coefficient. $A_{nl}$ is defined as an integral over the orientation of a molecule, i.e.,

$$
A_{nl} = \int_{-1}^{1} d\mu \mu^n \exp\left(-\frac{\hbar^2 \mu^2}{4MD_2\omega} + \frac{i\kappa\mu a}{2}\right) P_l(\mu), \tag{2}
$$

with $a = 0.74\AA$ the equilibrium separation distance of the D-D bond, $\mu$ the cosine of the inclination angle of the molecular axis from the $z$ axis of a reference Euclidean coordinate system, and $P_l$ the Legendre polynomial of order $l$. 

TABLE I. Intrinsic scattering cross-sections \( S_{J,J'} \) associated with different rotational transitions [6].

| \( S_{J,J'} \) | Even-J(Ortho) | Odd-J(Para) |
|---------------|-------------|-------------|
| Even-J        | \( a_{coh}^2 + \frac{3}{2} a_{inc}^2 \) = 6.687/4\( \pi \) | \( \frac{3}{2} a_{inc}^2 = 1.530/4\pi \) |
| Odd-J         | \( \frac{3}{2} a_{inc}^2 = 0.705/4\pi \) | \( a_{coh}^2 + \frac{3}{2} a_{inc}^2 = 6.102/4\pi \) |

The input parameter \( S_{J,J'} \) (in units of barns) in [1] for transitions between different rotational states is deduced and listed in Table I. Note here that only the incoherent scattering length \( a_{inc} \) of a bound nuclide contributes to the ortho(even \( J \))/para(odd \( J \)) conversion.

In a \( D_2 \) solid, the populations of the even-J(ortho state) and the odd-J(para state) are typically determined by the ortho/para population of the gas phase before the \( D_2 \) is frozen into the solid. The self-conversion between these two species in the solid phase into a thermal Boltzmann distribution is extremely slow compared with the time scale of experiment. For example, a room temperature equilibrium \( D_2 \) is a mixture of 67% ortho-\( D_2 \) and 33% para-\( D_2 \). The conversion rate in the solid is measured to be 0.06%/hr [2], requiring about 7 months to reduce the para content to 1.65% from 33%. On the other hand, relaxation to thermal distributions is rapid among ortho- and para- species themselves. Consequently, in low temperature circumstances relevant to super-thermal solid deuterium source (T<20K), only ground states (\( J = 0 \) for ortho; \( J = 1 \) for para) are present.

In the case of neutrons scattered off a low temperature crystal with well-defined lattice structures, harmonic solid correlation functions should be applied to the translational part of (1). Following the standard treatment of lattice dynamics [8], we perform a phonon expansion

\[
< \exp(-i\hbar \cdot r_I) \exp(i\hbar \cdot r_V(t)) >_{\text{trans}} = \exp(-2W(\kappa)) \exp\{\kappa \cdot u(\kappa) \cdot u_V(t)\} \times \exp\{2W(\kappa)\} [1 + \kappa \cdot u(\kappa) \cdot u_V(t) + O(\kappa \cdot u(\kappa) \cdot u_V(t)^2)]
\]

An overall Debye-Waller factor is extracted in front, and only the first two terms are left for discussion, yielding the zero and one phonon exchange processes, respectively.

Unlike conventional applications of elastic solid correlation functions, the first term in the phonon expansion (zero phonon term) coupled to molecular internal energy states not only gives rise to UCN energy transition (mainly upscattering), but overwhelms any phonon contributions when para-\( D_2 \) is present. The conversion of para- into ortho- molecules provides direct energy transfer to UCN. This scattering cross-section without phonon couplings has the simple form:

\[
\frac{d\sigma}{d\Omega}^{0 \text{ phonon}}_{J=1 \rightarrow 0} = \frac{3}{4} a_{inc}^2 \frac{k'}{k} e^{-2W(\kappa)} \times [4j_1^2(\frac{\kappa a}{2}) C^2(101; 00)]
\]

where \( j_1(\frac{\kappa a}{2}) \) is a spherical Bessel function of order one [9]. The increase of the neutron momentum is definite, i.e.,

\[
k' = \sqrt{2m_n \Delta E_{10} / h}.
\]

A conversion energy \( \Delta E_{10} \) of 7 meV gives \( k' \) a value of 1.84\( \times 10^{10} \) cm\(^{-1}\). The momentum transfer \( \kappa \) can be well-approximated by \( k' \) for UCN (\( k' >> k_{UCN} = 1.27 \times 10^{10} \) cm\(^{-1}\)), and the above differential cross-section is isotropic, making the integration of (3) straightforward.

A Debye-Waller factor originating from the uncertainty of positions of lattice sites, reduces the amplitude by a factor of 0.76. The temperature independent total cross section of UCN upscattering \( \sigma_{10} \) is calculated to be 31 barns. This is at least of an order of magnitude larger than the phonon annihilation cross-section in a 4K solid.

The rate of loss of UCN in the solid is

\[
\rho_{ucn} = \frac{w_{fi}}{V},
\]

\[
= \rho_{ucn}[\sigma_{10} v_{ucn} \rho'],
\]

where \( \rho' \) is the density of para-\( D_2 \), taken to be 3\( \times 10^{22} \) cm\(^{-3}\). The corresponding upscattering time \( \tau_{up} \) of UCN in a pure para-\( D_2 \) molecular solid is therefore

\[
\tau_{up} = \frac{1}{\sigma_{10} v_{ucn} \rho'},
\]

\[
= 1.5 \text{ msec}.
\]

For normal-\( D_2 \) which retains the room temperature equilibrium ortho/para ratio, the upscattering time due to the spin relaxation of para species is 4.6 msec.

To estimate the phonon upscattering rates, we approximate the \( D_2 \) hcp/fcc lattice as a cubic lattice to simplify the treatment of polarization anisotropies. The expression of the incoherent double differential cross-section involving one phonon exchange is

\[
\frac{d^2 \sigma}{d\Omega d\epsilon}^{\text{phonon}}_{J \rightarrow J'} = \frac{k'}{k} \frac{h^2 k^2}{2M_{D_2}} e^{-2W(\kappa)} S_{J,J'} (2J' + 1)
\]

\[
\times \sum_n \left( \frac{h}{2M_{D_2} \omega} \right)^n \frac{1}{n!} \sum_{J'' = J - J'} |A_{nl}|^2 C^2(JJ'1; 00)
\]

\[
\times Z(\epsilon_{ph}) \left\{ \begin{array}{ll} n(\epsilon_{ph}) + 1 & \text{if } \epsilon_{ph} > 0 \\ n(\epsilon_{ph}) & \text{if } \epsilon_{ph} < 0 \end{array} \right\}
\]

where the energy of phonon \( \epsilon_{ph} = \epsilon + \Delta E(J' \rightarrow J) + n\hbar \omega \), complying with the law of conservation of energy. Positive and negative values of \( \epsilon_{ph} \) correspond to single
FIG. 1. UCN upscattering cross-section vs. temperature of solid D$_2$. The one-phonon annihilation cross-section in an ortho-D$_2$ solid (solid curve) and in a para-D$_2$ solid (dashed curve) are plotted. The dashed-dagger line is the temperature independent UCN upscattering cross-section involving $J=1\rightarrow0$ relaxation not coupled to phonons in a para-D$_2$ solid.

phonon creation and annihilation, respectively. $Z(E)$ represents the normalized phonon density of states, and $n(E)$ the occupation number of phonons with energy $E$.

With a simple Debye model, in which

$$Z(E) = \frac{3E^2}{(k_BT_0)^3},$$

(7)

and the Debye temperature $T_0$ (110K for D$_2$) is the only parameter, a double integration of (7) with initial energy of UCN (see Figure 1) reproduces the upscattering cross-sections in ortho-deuterium calculated by Yu, Malik and Golub [4], in whose treatment rotational transitions were not considered. In an ortho-D$_2$ solid, the $J = 0 \rightarrow 0$ process dominates the upscattering. Even though the $J = 0 \rightarrow 1$ transition is energetically allowed through coupling to a phonon, the cross-section is kinematically suppressed by the smaller final phase space of upscattered neutrons. Para-D$_2$ has a distinguishably larger one-phonon annihilation cross-section than the ortho species. The origin of difference is again related to the $J = 1 \rightarrow 0$ relaxation channel. This provides UCN with additional energy to scatter into a larger volume of phase space, and secondly, it couples UCN to high energy phonons with large density of states, and is thus less restricted by the availability of phonon modes than the $J = 1 \rightarrow 1$ process. However, it is still suppressed by its small coupling to the phonon field, as opposed to the zero phonon term.

In summary, para-deuterium has a spin relaxation channel in which its conversion energy of 7 meV can be released to UCN, resulting in a temperature-independent short UCN lifetime of 4.6 msec in a normal-D$_2$ solid.

Elimination of the para-D$_2$ is necessary to achieve UCN lifetimes comparable to the nuclear absorption time in solid D$_2$.

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