Ultracold neutron upscattering rates in a molecular deuterium crystal

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A calculation of ultracold neutron (UCN) upscattering rates in molecular deuterium solids has been carried out, taking into account intramolecular excitations and phonons. The different molecular species ortho-D_2 (with even rotational quantum number J) and para-D_2 (with odd J) exhibit significantly different UCN-phonon annihilation cross sections. Para- to ortho-D_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_2 solid with 33% para content.

The low density of ultracold 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 β 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 superfluid ^{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_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 paper, we demonstrate that the presence of even very small concentrations of para-D_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 the same parity to preserve the symmetry of the wave function under permutation of identical particles. Young and Koppel[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 intermolecular degrees of freedom. The derived double-differential cross section in which the interference term is neglected has the form, for UCN with incident wave number k and final wave number k',

$$\frac{d^2\sigma}{d\Omega d\epsilon}=\frac{1}{2\pi\hbar}\frac{k'}{k}\int_{-\infty}^{\infty}dt\sum_l\langle\phi_{it}|e^{-i\kappa r_l(0)}e^{i\kappa r_l(t)}|\phi_{it}\rangle_{\mathrm{Trans}}$$
$$\times\sum_J P_{JS}\sum_{J'}\mathcal{S}_{JJ'}(2J'+1)e^{i(E_{J'}-E_J)t/\hbar}$$
$$\times\sum_{n=0}\frac{e^{in\omega t}}{n!}\left(\frac{\hbar^2\kappa^2}{2M_{D_2}\hbar\omega}\right)^n\sum_{l=|J'-J|}^{J'+J}|A_{nl}|^2$$
$$\times C^2(JJ'l;00), \qquad (1)$$

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, $E_J=7\ \mathrm{meV}\times J(J+1)/2$ is the rotational spectrum, $\hbar\omega$ is the intermolecular vibrational energy with n characterizing the number of vibrational energy quanta, and $C(JJ'l;00)$ is a Clebsh-Gordon coefficient. A_{nl} is defined as an integral over the orientation of a molecule, i.e.,

$$A_{nl}=\int_{-1}^{1}d\mu\mu^n\exp\left(-\frac{\hbar\kappa^2\mu^2}{4M_{D_2}\omega}+\frac{i\kappa a\mu}{2}\right)P_l(\mu), \qquad (2)$$

with $a=0.74\,\text{Å}$, the equilibrium separation distance of the D-D bond, μ 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.

The input parameter $S_{JJ'}$ (in units of barns) in Eq. (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 selfconversion between these two species in the solid phase into a thermal Boltzman distribution is extremely slow compared with the time scale of ex-

TABLE I. Intrinsic scattering cross sections $S_{JJ'}$ associated with different rotational transitions (Ref. 6).

$S_{JJ'}$	Even J (ortho-)	Odd J (para-)
Even J'	$a_{\rm coh}^2+5/8a_{\rm inc}^2=6.687/4\pi$	$3/4a_{\rm inc}^2=1.530/4\pi$
Odd J'	$3/8a_{\rm inc}^2=0.765/4\pi$	$a_{\rm coh}^2+1/4a_{\rm inc}^2=6.102/4\pi$

periment. 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%/h,[7] requiring about seven 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 superthermal solid deuterium source ($T<20$ K), 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 Eq. (1). Following the standard treatment of lattice dynamics,[8] we perform a phonon expansion

$$\begin{aligned}&\langle\exp\{-i\kappa r_l\}\exp\{i\kappa r_{l'}(t)\}\rangle_{\rm Trans}\\&\quad=\exp\{-2W(\kappa)\}\exp\{\langle\kappa u_l\kappa u_{l'}(t)\rangle\},\\&\quad=\exp\{-2W(\kappa)\}[1+\langle\kappa u_l\kappa u_{l'}(t)\rangle\\&\qquad+O(\langle\kappa u_l\kappa u_{l'}(t)\rangle^2)].\end{aligned}$$

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:

$$\left(\frac{d\sigma}{d\Omega}\right)_{J=1\to0}^{0\ \rm phonon}=\frac{3}{4}a_{\rm inc}^2\frac{k'}{k}e^{-2W(\kappa)}\left[4j_1^2\left(\frac{\kappa a}{2}\right)C^2(101;00)\right], \tag{3}$$

where $j_1(\kappa a/2)$ is a spherical Bessel function of order 1.[9] The increase of the neutron momentum is definite, i.e.,

$$k'=\sqrt{2m_n\Delta E_{10}}/\hbar. \tag{4}$$

A conversion energy ΔE_{10} of 7 meV gives k' a value of $1.84\times10^{10}\,{\rm m}^{-1}$. The momentum transfer κ can be well approximated by k' for UCN($k'\gg k_{\rm ucn}=1.27\times10^8\,{\rm m}^{-1}$), and the above differential cross section is isotropic, making the integration of Eq. (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[11] total cross section of UCN upscattering σ_{10} is calculated to be 31 b. This is at least of an order of magnitude larger than the phonon annihilation cross section in a 4 K solid.

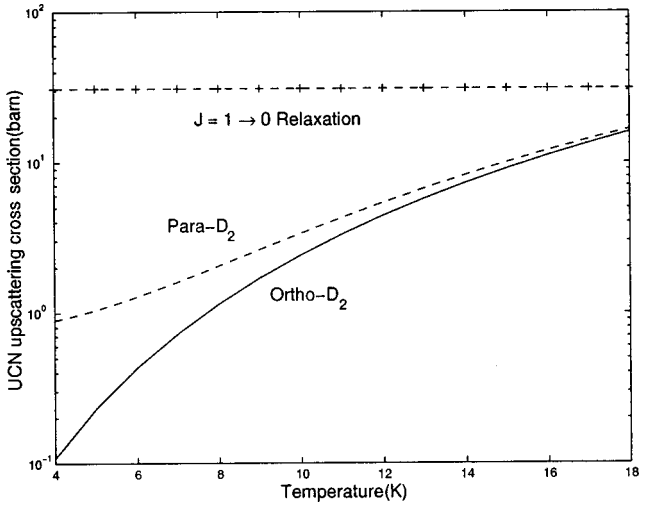

FIG. 1. UCN upscattering cross section vs temperature of solid D_2. The one-phonon annihilation cross section in an ortho-D_2 solid (solid curve) and in a para-D_2 solid (dashed curve) are plotted. The dashed-dagger line is the temperature independent UCN upscattering cross section involving $J=1\to0$ relaxation not coupled to phonons in a para-D_2 solid.

The rate of loss of UCN in the solid is

$$\dot\rho_{\rm ucn}=w_{fi}/V=\rho_{\rm ucn}[\sigma_{10}v_{\rm ucn}\rho'\,]. \tag{5}$$

Here ρ' is the density of para-D_2, taken to be $3\times10^{22}\,{\rm cm}^{-3}$. The corresponding upscattering time $\tau_{\rm up}$ of UCN in a pure para-D_2 molecular solid is therefore

$$\tau_{\rm up}=\frac{1}{\sigma_{10}v_{\rm ucn}\rho'}=1.5\ \ \text{msec}.$$

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 SD_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

$$\begin{aligned}\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{J\to J'}^{1\ \rm phonon}&=\frac{k'}{k}\frac{\hbar^2\kappa^2}{2M_{D_2}}e^{-2W(\kappa)}S_{JJ'}(2J'+1)\\&\quad\times\sum_n\left(\frac{\hbar\kappa^2}{2M_{D_2}\omega}\right)^n\frac{1}{n!}\\&\quad\times\sum_{l=|J'-J|}^{J'+J}|A_{nl}|^2C^2(JJ'l;00)\\&\quad\times\frac{Z(E_{\rm ph})}{E_{\rm ph}}\begin{cases}n(E_{\rm ph})+1 & \text{if } E_{\rm ph}>=0\\ n(E_{\rm ph}) & \text{if } E_{\rm ph}<0,\end{cases}\end{aligned} \tag{6}$$

where the energy of phonon $E_{\rm ph}=\epsilon+\Delta E(J'\to J)+n\hbar\omega$, complying with the law of conservation of energy. Positive and negative values of $E_{\rm ph}$ correspond to single 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

$$Z(E)=\frac{3E^2}{(k_B T_\Theta)^3}, \tag{7}$$

and the Debye temperature T_Θ (110 K for $D2$) is the only parameter, a double integration of Eq. (6) with initial energy of UCN (see Fig. 1) reproduces the upscattering cross sections in orthodeuterium 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 relatively 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, paradeuterium 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.

We have implemented a cryogenic para- to ortho-D_2 converter[7] which contains ferric-hydroxide, FeO(OH), powder as a catalyst. After flowing through the converter in which the catalyst is held at 17 K, the D_2 gas is converted predominantly to the ground (ortho-) state. The residual para- contamination was measured to be $1.4\%\pm 0.2\%$ by performing rotational Raman spectroscopy on the converted D_2 at room temperature.[10] Using this refined D_2 gas to prepare a solid UCN source, we have measured a prolonged UCN lifetime in this D_2 solid and an enhanced UCN production, both in agreement with predicted values.

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