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Ortho-para equilibrium in a liquid D_2 neutron moderator under irradiation

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The ortho-D_2 concentration of gas samples taken from the liquid D_2 cold moderator of the SINQ spallation neutron source at PSI has been measured. A value of $c_o = 0.762 \pm 0.001(\text{statistical}) \pm 0.006(\text{systematical})$ for the *in situ* quasiequilibrium at a temperature of 25 K and at an almost continuous power deposition of 230 ± 50 mW/g is established for samples taken after long irradiation periods. This unexpectedly low value, $c_o^{eq}(25\text{ K}) = 0.955$, is explained by a simple radiation-induced breakup and recombination model for D_2 molecules.

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I. INTRODUCTION

Nuclear exchange symmetry in the hydrogen molecules H_2, D_2 ($^2H_2$), and T_2 ($^3H_2$) leads to ortho and para states. States with higher multiplicity with respect to the nuclear spin are referred to as "ortho."[1,2] For H_2 and T_2, the total wave function is antisymmetric under exchange of the identical spin-1/2 nuclei, whereas the D_2 wave function is symmetric under exchange of the identical spin-1 deuterons. In D_2, nuclear wave functions for the spins $S=0$ and 2 are symmetric ($1+5=6$ magnetic substates, ortho) and require even angular momenta $J=0,2,\ldots$, whereas the $S=1$ states (3 substates, para) must have odd $J=1,3,\ldots$. In H_2 and T_2, the $S=0$ (para) spin wave function is antisymmetric and requires symmetric even J states, while $S=1$ (ortho) is coupled to odd J states. For all homonuclear hydrogens, the molecular ground states always have $J=0$ and the $J=1$ states are metastable.

Throughout the paper we use square brackets to denote number densities of atoms or molecules, e.g., $[D_2]=[D_2^o]+[D_2^p]$. The ortho-D_2 concentration is represented by c_o with $c_o=[D_2^o]/[D_2]$. For c_o in thermal equilibrium at a given temperature T we use $c_o^{eq}(T)$. There should be no danger of confusing tritium atoms T with temperature T.

The hydrogen systems have been investigated extensively (see Ref. 2 for a compilation), however, not much is known about the ortho/para (o/p) composition of liquid H_2 and D_2 cold neutron moderators. Under strong irradiation the equilibrium c_o will not necessarily correspond to $c_o^{eq}(T)$. The neutron flux obtained from a cold source is not very sensitive to its o/p composition, nevertheless, knowledge of the composition may be needed for designing optimized cold neutron moderators, performance calculations, and where interest is in maximizing fluxes of neutrons with the longest possible wavelengths. For application to very cold and ultracold neutron sources high ortho-D_2 concentrations are mandatory, otherwise severe depletion will occur through energy upscattering from the para- to ortho transition. However, see, e.g., Refs. 3 and 4 though for work on solid D_2. The present work is part of a program aimed at disclosing D_2 features relevant to the design of a new powerful source of ultracold neutrons which is under construction at the Paul Scherrer Institut (PSI).[5] The spallation neutron source at PSI, SINQ, includes a liquid D_2 moderator[6] and is, therefore, a source of liquid D_2 which has been subjected to strong irradiation. We present the results of an investigation of the ortho-D_2 concentration c_o of a sample taken from this source examining the c_o value in equilibrium, the time constant for reaching equilibrium, and its dependence on the radiation level.

II. EXPERIMENT

A. The SINQ cold moderator

Figure 1 shows a schematic arrangement of the SINQ cold moderator system. The proton beam (590 MeV, 1.2 mA) hits a heavy metal target causing spallation reactions that release fast neutrons which are thermalized in the room-temperature D_2O moderator. A 20-dm^3 liquid D_2 cold source is mounted in this moderator and produces cold neutrons for a neutron guide system and a conventional beam tube. A thermosyphon is used for the heat transfer between the source volume and the condenser system. It operates at about 150 kPa, giving a 25 K liquid temperature and a density of

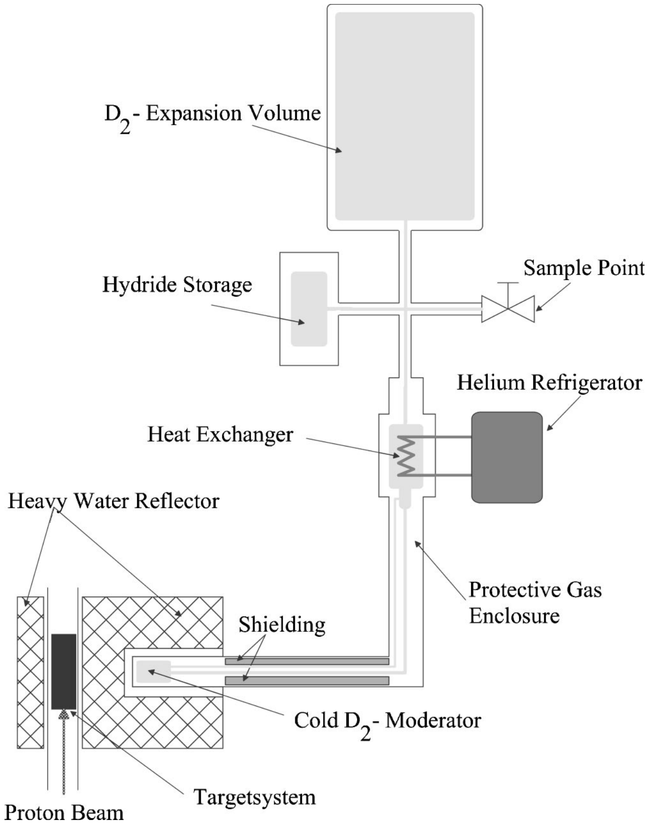

FIG. 1. Schematical view of the SINQ cold moderator facility (Ref. 6).

160 kg/m^3. The mass flow is about 5 $\mathrm{g\,s^{-1}\,mA^{-1}}$; about 1.5 dm^3/s of gas flowing from source volume to liquifier and 30 cm^3/s liquid in the return path. The system is usually filled once per year following an annual shutdown period of 2 to 3 months and hence begins with the deuterium with $c_o^{eq}(300\ \mathrm{K})\ (=2/3)$. At startup, the whole system (thermosyphon plus expansion volume) is filled with room-temperature D_2 gas at about 280 kPa (about 10 kg of D_2), then the liquifier is turned on and the thermosyphon is filled with liquid (about 3.5 kg). When operating with full power beam, medium energy, and fast neutrons and γ rays deposit a power of $190\pm40\ \mathrm{mW\,g^{-1}\,mA^{-1}}$ in the liquid D_2; the volume of liquid D_2 in the cold source chamber (19.8 dm^3) is reduced by about 1.5 dm^3 to give the pressure head to drive the thermosyphon. The 190 $\mathrm{mW\,g^{-1}\,mA^{-1}}$ power value comes from a performance calculation of the complete SINQ neutron source using the PSI version of the HETC code package.[7] Measured heat loads on the helium refrigerator, which includes contributions from both the D_2 and the container structure materials (550 and 120 W/mA, respectively, in the calculation), agree with the calculations to about 20%, leading to the assigned uncertainty.

B. Gas sampling and analysis

The c_o measurement of the irradiated liquid D_2 is obtained from the analysis of gas samples. The sample on which the present work is based was taken after irradiation during a full SINQ cycle from April to December 2002. The proton beam availability on the SINQ target was about 85% between September and December, dominated by the regular service shutdowns of one day per week. The sample was taken immediately after shutting down the proton beam; the refrigerator system of the cold source was switched off and the D_2 cover gas removed from the gas system between the sample point and the liquid by use of the hydride storage system; this in order to prevent admixtures of gas with different c_o. A 40-cm^3 gas sample container (stainless steel 304L) was filled with D_2 gas at 190 kPa. Within half an hour after sampling, D_2 from this container was transferred into a spectroscopy cell of about 2.5 cm^3 volume made from Al with four glued sapphire windows.

The c_o values were measured by means of frequency resolved Raman spectroscopy. The Raman cross section involves the isotropic (α') and anisotropic (γ') invariants of the Raman polarizability tensor. Values for D_2 spectroscopy can be found in, e.g., Ref. 8. The rotational dependence of the cross section is usually represented by Placzeck-Teller coefficients and is negligible in isotropic Raman measurements.[9] Hence, the degeneracy of rotational states due to nuclear-spin substates becomes apparent in the fractional population of the rotational states, and the intensity ratio of odd and even rotational Raman lines is a linear and direct measure of the o/p concentration ratio. The rotational quantum numbers J remain unchanged in the observed Q-branch transitions to the first vibrational state and the corresponding lines are resolved by our spectrometer system (70-mW Ar ion laser, SPEX14018 (3/4)-m double-grating spectrometer, 1800 lines/mm, charge-coupled device readout). Corrections due to laser polarization and the polarization dependent reflection efficiency of the spectrometer gratings have to be taken into account and are cross checked with the known value $c_o^{eq}(300\ \mathrm{K})=2/3$ for D_2 gas at room-temperature equilibrium. Figure 2 shows three vibrational Raman spectra: (i) for D_2 from a factory cylinder, $c_o^{eq}(300\ \mathrm{K})=2/3$; (ii) for D_2 catalyzed at the triple point using OXISORB® as a catalyst,[10,11] $c_o=c_o^{eq}(18.7\ \mathrm{K})=0.985$; and (iii) for the D_2 sample taken from the SINQ moderator.

The triple point catalyzed D_2 gas, with $c_o=0.985$, was used to measure the time constants for o/p relaxation in the sample container and the spectroscopy cell. Values of 36 and 8 days, respectively, were obtained, hence only small relaxation corrections are needed in order to extrapolate back to the c_o value at the time of sampling.

C. Results

The Q branch $(\Delta J=0,\ \Delta\nu=1)$ rovibrational Raman spectrum for the sample from SINQ shown in Fig. 2 is the sum of two measurement cycles, each 100 min long and on average 12 h after sampling. The analysis of the fit to the line intensities yields $c_o=0.756\pm0.001\pm0.004$; the first uncertainty is due to counting statistics and the second one due to systematics introduced by different possible background models. The correction for relaxation in the sample cell using the measured time constant results in an increase of the mea-

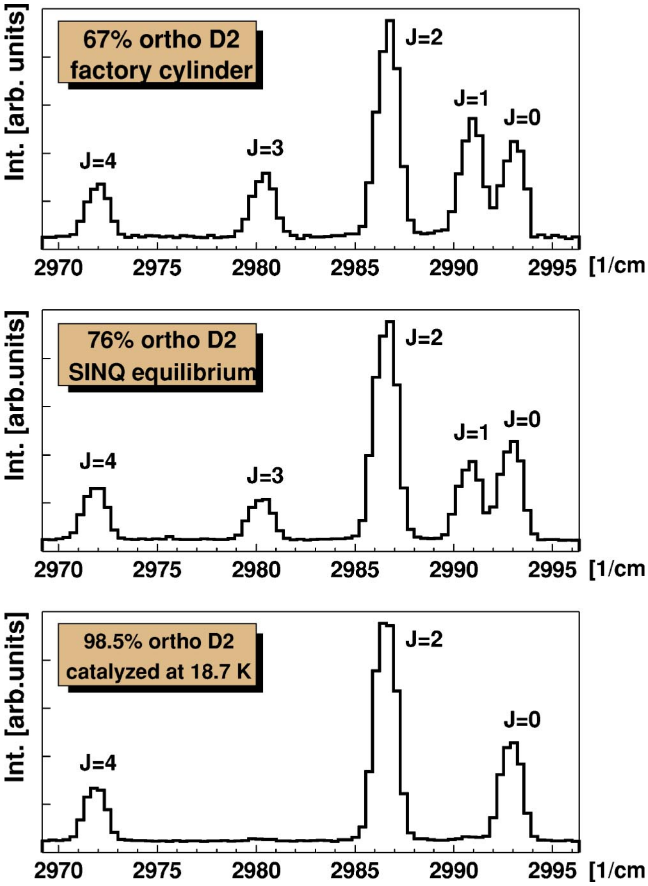

FIG. 2. Intensities (normalized to the even J intensity) as a function of the wave number for the Q branches of three different D_2 samples: at room-temperature equilibrium ($c_o=0.67$), from SINQ ($c_o=0.76$) and converted at 18.7 K ($c_o=0.98$).

sured value by 0.006±0.004. Another systematic uncertainty is introduced by the different vapor pressures of liquid ortho and para D_2. Using vapor pressure curves as given by Ref. 2 we estimate the shift to be ± 0.002 at maximum. The final result thus reads $c_o=0.762\pm 0.001(\text{statistical}) \pm 0.006(\text{systematical})$.

We note the accidental numerical coincidence of the simple mixture of two-third D_2 with $c_o=2/3$ from the expansion tank with one-third D_2 at $c_o^{eq}(25\text{ K})=0.955$, yielding $(2/3)(2/3)+(1/3)(0.955)=0.763$. However, this process is excluded by the sampling technique. A mechanism that could explain that the lower c_o value is a continuous feeding of D_2 gas with a larger para content into the cold system; this is excluded by the very slow diffusion in the gas system (the diffusion time constant of D_2 between the liquid and the expansion volume through about 60 m long, 90 mm diameter tubing is estimated to be of the order of one year) and the very long relaxation times (order months) for D_2 gas evaporating from the liquid.

III. MODEL CALCULATIONS

For the interpretation of the measured results, we adopt a simple model (as, e.g., used in Ref. 12) in which the only relevant processes are molecular breakup leading to free-atom production under irradiation, recombination of atoms to molecules, and o/p conversion. More complex chemical processes including, e.g., reactive ions ($D^+, D_3^+, \ldots$) are neglected:

$$\left.\begin{array}{l} D_2^o \xrightarrow{\;k\;} \\ D_2^p \xrightarrow{\;k\;} \end{array}\right\} 2\,D \left\{\begin{array}{l} \xrightarrow{2\alpha/3} D_2^o \\ \xrightarrow{1\alpha/3} D_2^p \end{array}\right. \tag{1}$$

$$D_2^p \xrightarrow{\beta c_o^{eq}(T)} D_2^o \xrightarrow{\beta(1-c_o^{eq}(T))} D_2^p. \tag{2}$$

Here, k denotes the molecular breakup rate due to irradiation and α the recombination rate. For recombined D_2, assuming high-temperature o/p equilibrium (2:1), leads to ortho- and para-D_2 production rates due to recombination of $2\alpha/3$ and $1\alpha/3$, respectively. The conversion rate β pulls towards thermal equilibrium $c_o^{eq}(T)$ at a given temperature T [e.g., $c_o^{eq}(25\text{ K})=0.955$].

A. Free atom formation under irradiation

The molecular breakup rate k is obtained from the known power deposition of $190\pm 40\text{ mW g}^{-1}\text{ mA}^{-1}$ in the liquid D_2 of the cold source. The major part of the energy deposition is due to neutrons and photons which in secondary processes contribute to ionization and molecular breakup. Assuming an energy of 36.6 eV for the creation of an ion pair and 5.1 atoms created per ion pair[2,12] and $(1.5\times 10^{23})\text{-g}^{-1}$ D_2 molecules one obtains

$$k=(5.5\pm 1.1)\times 10^{-7}\text{ s}^{-1}\text{ mA}^{-1}. \tag{3}$$

Obviously, this molecular breakup rate is valid only for beam-on periods. However, beam-off periods (one- or two-day shutdowns of the accelerator as well as short breaks) cannot be treated by a reduced average molecular breakup rate because the recombination takes place on a much shorter time scale than these interruptions. The unirradiated conversion is slow compared to the radiation-induced process, thus beam-off times can be neglected in a first approximation for the equilibrium c_o and only stretch the time needed to obtain equilibrium. A similar argumentation is valid for D_2 which is circulating round the thermosyphon during beam-on times. Because the recombination rate is much larger than the matter exchange rate between cold moderator volume and heat exchanger volume, the relevant conversion processes take place in the moderator and the nonirradiated fraction of the volume only leads to a dilatation of the equilibration time.

B. Recombination of atoms to molecules

The recombination rate α depends on the diffusion constant of free atoms in liquid D_2 and quadratically on the number of available atoms. In principle, it can be measured in experiments where the free-atom concentrations and the temperature are varied. This has been done in the case of solid hydrogens.[13,14] Also, α could be obtained by direct theoretical calculation. However, as will become clear below, we follow another approach which allows us to relate a function of α and k to our measured c_o value without actually determining α.

C. Ortho/para conversion

The conversion between the ortho and para states strongly depends on environmental conditions and one has to consider four different contributions, of which only the first two turn out to be relevant in our application, given as follows:

(i) The "natural" conversion due to molecule-molecule interactions can have two different contributions (one usually neglects the thermally hindered channel in which para D_2 is created in the collision of two ortho-D_2 molecules, see below)

$$\frac{d}{dt}[D_2^o]=\beta_1[D_2^o][D_2^p]+\beta_2[D_2^p]^2 \tag{4}$$

$$=\beta_1[D_2^o]([D_2]-[D_2^o]) + \beta_2([D_2]-[D_2^o])^2 \tag{5}$$

$$=(\beta_1-2\beta_2)[D_2][D_2^o] - (\beta_1-\beta_2)[D_2^o]^2+\beta_2[D_2]^2. \tag{6}$$

Experimental and theoretical evidence exist for the two rate constants β_1 and β_2 being equal, which reduces Eq. (6) to a first-order differential equation[15]

$$\frac{d}{dt}[D_2^o]=\beta_{12}[D_2]([D_2]-[D_2^o]). \tag{7}$$

The time constants for the natural conversion in liquid D_2 are of the order of 3100 h (130 days),[2,16] corresponding to $\beta_{12}[D_2]\approx 0.9\times 10^{-7}\,\mathrm{s}^{-1}$. However, due to effectively neglecting the para production, $c_o=1$ is obtained for long times. In order to correct for this behavior, we slightly modify Eq. (7) to obtain

$$\frac{d}{dt}[D_2^o]=\beta_{12}\;c_o^{eq}(T)[D_2]^2-\beta_{12}[D_2][D_2^o]. \tag{8}$$

Equation (8) preserves the experimentally observed first-order behavior and yields the correct long-time limit $[D_2^o]\to c_o^{eq}(T)[D_2]$.

(ii) The conversion in collisions of free D-atoms (produced under irradiation) with molecules (compare Ref. 12)

$$\frac{d}{dt}[D_2^o]=\beta\;c_o^{eq}(T)[D][D_2^p]-\beta(1-c_o^{eq}(T))[D][D_2^o] \tag{9}$$

$$=\beta c_o^{eq}(T)[D][D_2]-\beta[D][D_2^o]. \tag{10}$$

(iii) The catalytic conversion in collisions with wall material (effective surface area A) is, except when c_o is very close to $c_o^{eq}(T)$, a zero-order reaction with time constant β_3,

$$\frac{d}{dt}[D_2^o]=\beta_3 A. \tag{11}$$

Typical values for good catalysts are of the order of $10^{-6}\,\mathrm{mol/m^2\,s}$.[2] If we assume the cold moderator wall material to be a good catalyst with a surface area of the order of 1 $\mathrm{m^2}$ or less, and we deal with the order of 10^3 mol D_2, conversion times of the order of 10^9s are obtained. One would need catalytic areas of 100 $\mathrm{m^2}$ in order to get rates comparable with natural conversion. In SINQ, the wall material of the moderator and the heat exchanger is aluminum and the system is rather clean, thus, we consider this contribution to be negligible.

(iv) The catalytic conversion in collisions with paramagnetic impurities (such as oxygen molecules) is a first-order process (compare, e.g., Ref. 17):

$$\frac{d}{dt}[D_2^o]=\beta_4\;c_o^{eq}(T)[O_2][D_2]-\beta_4[O_2][D_2^o]. \tag{12}$$

From admixtures of $[O_2]=(10^{-4}\text{–}10^{-3})[H_2]$ to solid H_2, a value $\beta_4=1.15(8)\,\mathrm{s}^{-1}$ for the high-temperature limit (above 8 K, not limited by molecular diffusion, but by the actual conversion process) was obtained.[17] This should not be very different in liquid H_2 and in liquid D_2. However, the solubility of O_2 in liquid H_2 (and thus in D_2) is rather small and estimated to be of the order or less than $[O_2]\approx 10^{-8}[H_2]$.[2] Thus $\beta_4[O_2]\approx 10^{-8}\,\mathrm{s}^{-1}$, which would amount to only a small correction to the natural self-conversion and can be neglected here.

D. The resulting equation and its solution

Combining the relevant processes, Eqs. (1), (2), (8), and (10), one gets:

$$\frac{d}{dt}[D_2^o]=-(k+\beta[D]+\beta_{12}[D_2])[D_2^o]+(\beta[D] + \beta_{12}[D_2])c_o^{eq}(T)[D_2]+\frac{2}{3}\alpha[D]^2. \tag{13}$$

The first solution of Eq. (13) is obtained by treating SINQ as a continuous, steady-state source; this is a valid approximation because, as may be seen below, the time constants for ortho-para conversion are long (weeks) compared to typical beam interruption times (seconds, minutes, sometimes days). Obviously, for the steady state, the rate of free-atom production must equal the recombination rate:[18]

$$\frac{d}{dt}[D]=2k[D_2]-2\alpha[D]^2\overset{!}{=}0 \tag{14}$$

yielding

$$[D]=\sqrt{\frac{k}{\alpha}[D_2]}, \tag{15}$$

which allows us to define $\beta^*=\beta\sqrt{[D_2]/\alpha}$,

$$\beta[D]=\beta^{*}\sqrt{k} \tag{16}$$

Inserting $\alpha[D]^2$ from Eq. (14) into Eq. (13) and using Eq. (16) yields

$$\frac{d}{dt}[D_2^o]=-(k+\beta^{*}\sqrt{k}+\beta_{12}[D_2])[D_2^o]+\left(\beta^{*}\sqrt{k}c_o^{eq}(T)+\beta_{12}[D_2]c_o^{eq}(T)+\frac{2}{3}k\right)[D_2]. \tag{17}$$

For constant k, Eq. (17) is easily solved as

$$[D_2^o]=C\exp\{-(k+\beta^{*}\sqrt{k}+\beta_{12}[D_2])t\}+\frac{\left\{\beta^{*}\sqrt{k}c_o^{eq}(T)+\beta_{12}[D_2]c_o^{eq}(T)+\frac{2}{3}k\right\}[D_2]}{k+\beta^{*}\sqrt{k}+\beta_{12}[D_2]} \tag{18}$$

in which the integration constant C is determined by the initial conditions at $t_0=0$:

$$C=\ c_o(t_0)-\frac{\left\{\beta^{*}\sqrt{k}c_o^{eq}(T)+\beta_{12}[D_2]c_o^{eq}(T)+\frac{2}{3}k\right\}[D_2]}{k+\beta^{*}\sqrt{k}+\beta_{12}[D_2]}. \tag{19}$$

For long times this leads to the expected steady-state behavior. The steady state is approached with the time constant $\tau=1/(k+\beta^{*}\sqrt{k}+\beta_{12}[D_2])$ which for $k\gg\beta,\beta_{12}$ is only slightly slower than $1/k$.

Considering the steady state, $d/dt[D_2^o]=0$, one obtains from Eq. (17),

$$c_o=\frac{[D_2^o]}{[D_2]}=\frac{\beta^{*}\sqrt{k}c_o^{eq}(T)+\beta_{12}[D_2]c_o^{eq}(T)+\frac{2}{3}k}{k+\beta^{*}\sqrt{k}+\beta_{12}[D_2]} \tag{20}$$

which naturally yields the two limiting cases

$$c_o=c_o^{eq}(T)\quad\text{no irradiation,}\quad k=0 \tag{21}$$

and

$$c_o=\frac{2}{3}\quad\text{for}\quad k\gg\beta,\beta_{12}. \tag{22}$$

IV. SPECIFIC RESULTS FOR SINQ AND DISCUSSION

The measured value for c_o, the estimated value for k [Eq. (3)], and a constant proton beam current of 1.2 mA on the SINQ target are used to obtain a value for $\beta[D]$ $(=\beta^{*}\sqrt{k})$ from Eq. (20):

$$\beta[D]=\frac{(c_o-2/3)k-(c_o^{eq}(T)-c_o)\beta_{12}[D_2]}{c_o^{eq}(T)-c_o}=(2.36\pm0.64)\times10^{-7}\,\mathrm{s}^{-1} \tag{23}$$

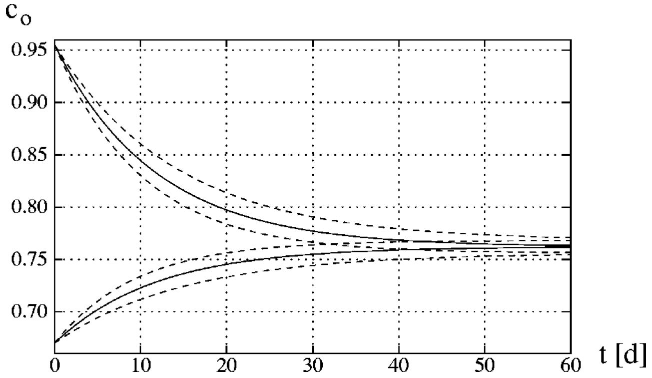

FIG. 3. Calculated temporal development of the ortho-D_2 concentration [Eqs. (18) and (19)], assuming 1.2-mA continuous proton beam and starting either at room-temperature equilibrium ($c_o=2/3$) or at thermal equilibrium [$c_o^{eq}(25\ \mathrm{K})=0.955$]. The dashed error bands indicate the possible range, simply adding the uncertainties in the power deposition and in c_o.

which then gives [Eq. (16)]

$$\beta^{*}=(2.91\pm0.55)\times10^{-4}\sqrt{\mathrm{s}^{-1}}. \tag{24}$$

These values are used in Eq. (17) to calculate the temporal development of $[D_2^o]$. Results starting from either $c_o(t_0)=2/3$ or $c_o(t_0)=0.955$, respectively, are shown in Fig. 3. After a few weeks of operation the system has come into an equilibrium independent of the initial concentrations.

The influence of proton current variation has been investigated directly by solving Eq. (17) using the measured values. The proton beam onto SINQ is logged every minute and the profile for the period April to December 2002 is shown in Fig. 4. The temporal development of c_o obtained from Eq. (17) is displayed in the lower part of Fig. 4.

The calculation uses $c_o(t_0)=2/3$ but other initial values lead to exactly the same behavior after less than 2 months. At two intermediate dates (June 19 and September 4) about half of D_2 was evaporated from the cold moderator for sampling tests. For the calculation we assumed complete mixing of the cold D_2 with the buffer gas at room-temperature equilibrium. Again, any initial o/p ratio at these dates returns to the same c_o on the 23rd of December.

Using the result for β^{*} obtained from the SINQ sample [see Eq. (24)] the dependence of c_o on k is calculated from Eq. (20). The result is shown in Fig. 5.

Qualitatively, one can discuss the behavior at higher power densities than in SINQ by using results for tritium. The self-heating due to β-decay amounts to 1 W per mol of tritium atoms giving a $k_{T_2}\approx1.4\times10^{-6}\,\mathrm{s}^{-1}$ (note $k=2.84\times10^{-6}\,\mathrm{s}^{-1}$ is given in Ref. 12 for the atom formation rate in agreement with our molecular breakup rate k_{T_2}). In T_2 no obvious discontinuity exists in the ortho-para transition rate at the solid-liquid phase transition;[19] this suggests that some liquid T_2 properties can be extrapolated from solid T_2 data. The ortho fraction in liquid T_2 can thus be expected to be $\geqslant0.6$. Thich is consistent with the finding that in liquid D_2 the hot limit is not far beyond the SINQ data point.

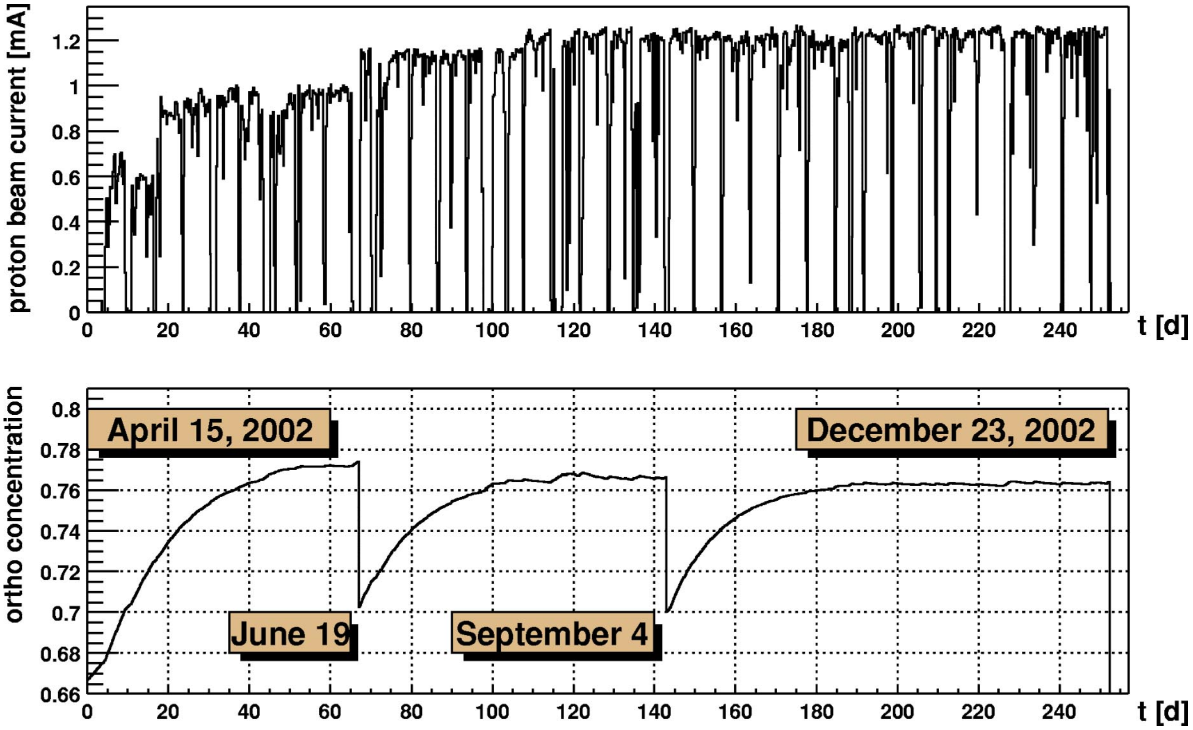

FIG. 4. The upper figure displays the proton beam current on the SINQ target over year 2002. Using this as an input, the lower graph shows the development of the ortho-D_2 concentration as calculated from Eq. (17), starting arbitrarily at $c_o = 2/3$ and using the values for k and β^* which were obtained in Eqs. (3) and (24).

Using our model, one can also predict c_o values for the two liquid D_2 moderators at the Institute Laue-Langevin (ILL). The relevant power depositions are 5.5 kW in 25-dm^3 D_2 (ILL 1) and 3 kW in 6 dm^3 (ILL 2), respectively.[20] On the basis of the power distribution in the SINQ cold source, we use 80% of these values as the power in the liquid D_2 and deduce approximate breakup rates $k(\mathrm{ILL1}) \approx 2.5 \times 10^{-6}\ \mathrm{s}^{-1}$ and $k(\mathrm{ILL2}) \approx 5.8 \times 10^{-6}\ \mathrm{s}^{-1}$. Using these values in Eq. (20) one obtains $c_o(\mathrm{ILL1}) \approx 0.72$ and $c_o(\mathrm{ILL2}) \approx 0.70$.

An experiment with a 6 dm^3 volume D_2 cold source in the VVRM reactor at PNPI Gatchina[3,21] reported no effect in solid D_2 and a fast o/p conversion to $c_o = 0.95 \pm 0.05$ in liquid D_2 at a power deposition of 11 ± 1 mW/g ($k \approx 3 \times 10^{-8}\ \mathrm{s}^{-1}$ correspondingly). Our model reproduces this value but we believe this to be a coincidence: We expect a slow equilibration time dominated by the natural conversion. Liquid and solid tritium data[12,19,22] show that the conversion rate of the (self-)irradiated hydrogen species increases when the temperature is lowered and reaches a maximum for T_2 at 11.4 K.[22]

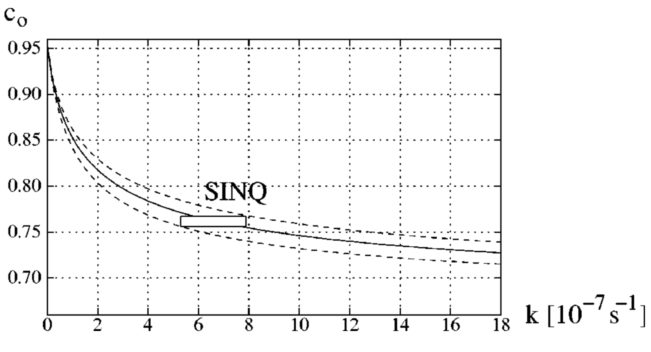

FIG. 5. The dependence of the equilibrium ortho-D_2 concentration c_o at 25 K on the molecular breakup rate [see Eq. (20)], which is proportional to the radiation power deposition in the liquid D_2. The uncertainties are dominated by systematics and have been added linearly.

V. CONCLUSIONS AND OUTLOOK

We have presented results of a measurement of the ortho-D_2 concentration in the liquid D_2 moderator of the SINQ spallation neutron source at PSI. The measured value $c_o = 0.762 \pm 0.001(\mathrm{statistical}) \pm 0.006(\mathrm{systematical})$ is far from thermal equilibrium at 25 K, but can be understood in terms of irradiation-induced breakup and recombination of D_2 molecules. The model used in order to interpret the result yields c_o as a function of the breakup rate. Generally, stronger irradiation causes higher conversion rates and leads to reduced c_o values. The values for tritium, which has a very strong self-irradiation, fits within this picture. Extrapolation to lower irradiation values is difficult due to the increased importance of other conversion mechanisms (see Sec. III C). More data points would be interesting, such as with different proton beam currents on the spallation neutron source or sampling from other moderators, e.g., the liquid D_2 cold moderators of the ILL Grenoble reactor. With respect to solid D_2 moderators for ultracold neutron sources, one can conclude that radiation assisted conversion in the liquid state is no viable option for achieving high c_o values. However, the radiation dependent c_o equilibrium, together with the atom mobility, is a strong function of temperature. In solid D_2 at low temperatures conversion is favored over recombination, yielding large c_o equilibrium values. If the radiation field is too weak to only rely on radiation assisted conversion in solid D_2, a suitable catalyst needs to be used to prepare $c_o = c_o^{eq}(18.7\ \mathrm{K}) = 0.985$. Starting from this value one can then achieve even higher c_o values due to irradiation as well as due to natural conversion.

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