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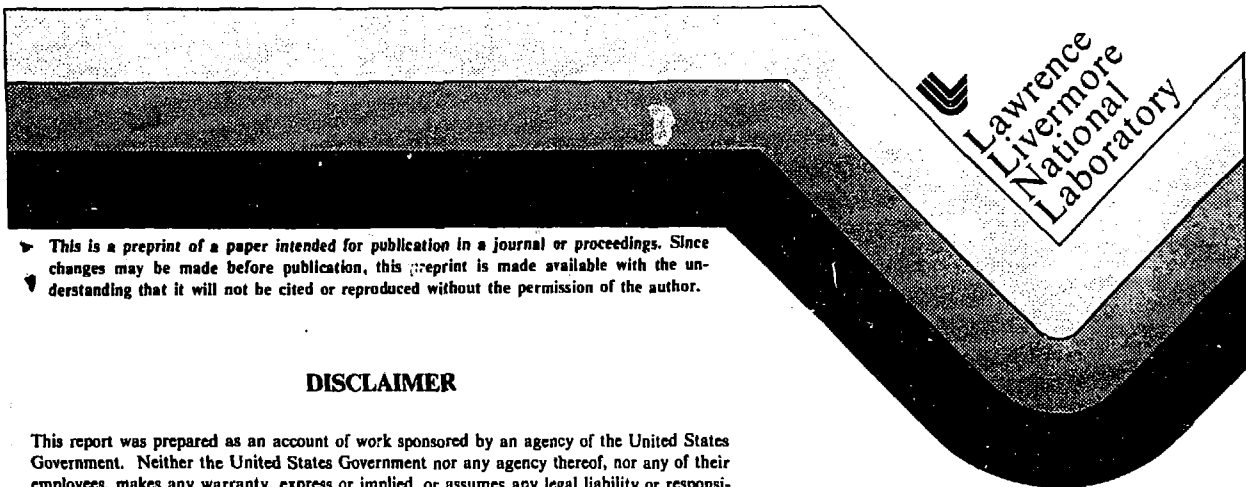
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ABSTRACT

The expected value of nuclear spin polarization to inertial confinement fusion is recapitulated. A comparison of brute force versus dynamic nuclear polarization, as applied to solid deuterium-tritium, is given, and the need for a long triton polarization memory time (longitudinal nuclear relaxation time) is shown. The time constant for 25 mol% T_2 -50 DT-25 D_2 is a short 0.3 s at 5 K and waiting lowers it to 0.1 s. Use of 90 to 96 mol% molecular DT raises the time constant to 0.9 s and addition of about 20 mol% nH_2 increases it to 5 to 7 s. The theory shows that the species shortening the triton memory time is the $J=1 T_2$, which can be reduced in our samples only by self-catalysis. The heating in order to mix in nH_2 increases the percent of $J=1 T_2$ and mixing may not be perfect. The experiments have increased the triton memory time twenty-fold and shown that removal of the $J=1 T_2$ is the key to improved results.

Introduction

One of the truly original ideas in controlled hydrogen fusion is nuclear spin polarization. Kulsrud, Furth, Valeo and Goldhaber, applied this idea to deuterium-tritium (D-T), the fusion fuel of lowest ignition temperature.^{1,2} That the cross section of a nuclear reaction can depend on the nuclear spin was shown in 1934 by Goldhaber.³ In the case of D-T, the deuteron and triton form an intermediate complex state of He⁵, which has a resonant state at 107 keV above the energy of the free reactant nuclei.⁴ This has a nuclear spin of 3/2, which can be obtained only by directly adding the spin-1/2 triton to the spin-1 deuteron, i.e. the D-T must be polarized with their nuclear spins parallel to each other.

For totally spin-polarized D-T, the fusion cross section is expected to rise 50% over the random D-T mix.¹ Pan and Hatchett have shown that such a material could reduce the input energy requirement of an inertial confinement fusion (ICF) driver by almost a factor of two.⁵ More has calculated that the intense laser beam of the driver will not destroy the polarization of the target, because because the irradiation time is so short.⁶ Cicchitelli, et. al., postulate a gain of three by using polarized D-T pellets in magnetic fusion reactors.⁷

Of interest to us is how to polarize solid D-T, which will require temperatures of 4 K or below. A method sure to succeed is "brute-force" whereby the sample is placed in a large DC magnetic field at millikelvin temperatures. Honig has successively done this with solid HD.⁸ However, the tritium radioactivity, which delivers 1 W/mol in D-T, makes it unlikely that ultralow temperatures can ever be reached, except in

vanishingly small samples. We suggested using instead dynamic nuclear polarization (DNP).⁹ This is the method employed in almost all polarized targets for nuclear physics. Here, one uses the hydrogen atoms formed in the solid by the radiation. Each atom possesses an unpaired electron, whose large magnetic moment may be easily polarized in a DC magnetic field at 1 to 4 K. Microwave energy of almost the electron spin resonance (e.s.r.) frequency is then added. If conditions are right within the sample, the electrons will transfer their polarization to the nuclei. There are two oppositely-behaved mechanisms for DNP. One is called the solid state (or solid) effect, and it works only for a narrow e.s.r. line.^{10,11} The other is called thermal mixing (or dynamic cooling), and it requires a broad e.s.r. line.¹²⁻¹⁴ It is not clear which mechanism will prevail in solid D-T.

The two theories of DNP agree on one thing: the nuclei must have a polarization memory time, T_1 (called in nuclear magnetic resonance (n.m.r.) the longitudinal nuclear relaxation time). It must be long because: 1) in DNP, each atom must polarize hundreds or thousands of nuclei in succession, and 2) time is needed after polarization to perform the ICF shot. In general, T_1 for solid hydrogen is short, as we shall see. The triton has a larger magnetic moment than the deuteron and hence a shorter relaxation time. More effort has, therefore, centered on the triton.

Much may be learned by considering the proton, which is magnetically similar to the triton. Honig greatly lengthened T_1 for the proton in solid HD by waiting at cryogenic temperatures for the $J=1$ H_2 (first metastable excited rotational state) to slowly de-excite to $J=0$ H_2 .¹⁵ He found that radiation lowered T_1 .¹⁶

Another approach is needed. One such approach is to cool to 0.3 K in a dilution refrigerator - the usual technique in DNP. While this may yet be necessary, any attempt to operate with the simpler 1-to-4 K cryogenic technology can only benefit this inherently difficult program. Also, starting with a high 4 K T_1 value is likely to produce an even higher one upon cooling. The approach we shall take here is to try to manipulate the polarization memory time, T_1 , at liquid helium temperature.

Theory of the Polarization Memory Time

In order to understand the nuclear relaxation of solid hydrogen which is not isotopically pure, we need a theory. Such a theory was constructed by Moriya and Motizuki for the case of $J=0$ and $J=1$ D_2 ,¹⁷ and it has been elaborated on by Gaines and Souers.¹⁸ The theory is expected to hold from 1.2 to about 10 K. At lower temperatures the theory may still hold, but it has not been checked. Above 10 K, the additional mechanism of molecular diffusion appears.

For a general solid hydrogen, we may write the total relaxation time, $T_1(\text{total})$, as the sum of three mechanisms.

$$\frac{1}{T_1(\text{total})} = \frac{1}{T_1(\text{sl})} + \frac{1}{T_1(\text{p})} + \frac{1}{T_1} \quad (1)$$

Here, $T_1(\text{sl})$ is the spin-lattice relaxation time, which is very long in solid hydrogen and does not matter.¹⁹ The quantity $T_1(\text{p})$ is a paramagnetic relaxation time that should occur only in an irradiated sample with many paramagnetic defects. It has not been yet seen but is expected. This leaves T_1 as the mechanism of greatest importance and the one we will

consider here at length. It is actually a spin-rotation time constant. The J=1 molecules in the solid possess electric quadrupole moments, which interact to split the rotational energy level of the J=1 state into a band. When the difference of states within the band equal the n.m.r. frequency, energy freely flows from the nuclei to the molecular rotational degree of freedom. All the nuclear magnetic energy must flow through the nuclei of the J=1 species it is interacting with. The less J=1 hydrogen is present, the greater is the bottleneck and the longer T_1 will be. This produces the long times seen in solid HD with very low J=1 H_2 content.

Let us consider the proton in solid HD. Its nuclear resonance functions independently of the deuteron in the same molecule. Its relaxation time will be

$$T_1(\text{H in HD}) = \frac{[\text{HD}](1/2)(3/2) + [\text{J=1 } H_2](1)(2)}{[\text{J=1 } H_2](1)(2)} T_{11} \quad (2)$$

The brackets are the mol percent of the given species. The numbers following are the nuclear spin factors $I(I+1)$, where I is the nuclear spin: 1/2 for H in HD and 1 for H in J=1 H_2 . The product of composition times spin yields the total nuclear energy. Eq. 2 says that all the nuclear magnetic energy of the protons in the numerator must funnel through the nuclear energy "gateway" to the J=1 molecular rotation in the denominator. The term T_{11} is the "inherent" relaxation time, and it equals T_1 for pure H_2 . It possesses a minimum at about 1 mol% of J=1 H_2 but is larger for smaller or larger J=1 values. This minimum is the unfortunate result of the particular fluctuation spectrum of the J=1 quadrupole moment. This fluctuates from a zero value to some cut-off frequency and acts like a "resonance" in interacting with the nuclear magnetic system.

It is important to realize that only J=1 H₂ figures in the denominator of Eq. 2 but all J=1 species add into the calculating of T₁₁. For example, T₁₁ will depend on whatever J=1 D₂ was present in addition to J=1 H₂.

In Eq. 2, nH₂ is usually used to do the experiment, so that one part of inert J=0 H₂ exists for every three parts of J=1 H₂. The amount of HD is 100% less the two types of hydrogen. Because there is no radioactivity, the solution is stable indefinitely at cryogenic temperatures. Eq. 2 becomes

$$T_1(\text{H in HD}) = \frac{75 + [J=1 \text{ H}_2]}{2[J=1 \text{ H}_2]} T_{11} \quad (3)$$

We summarize all comparable proton data for solid H₂ and HD in Table 1.²⁰ The data was taken at 4 K, but we expect it to be little changed over the 2 to 10 K region. The relaxation time is expected to be a function of the n.m.r. frequency, so we list the experimental frequency used. It is upon this table that the validity of the theory rests. The H₂ data was measured by Meyer^{21,22} and Hardy and Gaines.²³ The HD data was taken by Hardy and Gaines²⁴ and by Honig.¹⁵ All the columns are directly comparable except the farthest right, which is an example of the measured HD data that T₁₁ is calculated from. All HD contains a small amount of J=1 D₂ which will affect T₁ but cannot be accounted for in these experiments. The T₁₁-minimum is evident just under 1%.

The important result of Table 1 is that T_{11} for H_2 (actually, T_1) and for HD are the same, thereby confirming the theory.

We now turn to the solution D_2 -DT- T_2 , where D_2 and T_2 each come in the $J=0$ and $J=1$ forms but DT is always $J=0$ at cryogenic temperatures. The triton is almost identical in its nuclear magnetic properties to the proton, and DT is, therefore, almost identical to HD. We may carry over the theory with almost no change and obtain:

$$T_1(\text{T in DT}) = \frac{3/4 \{ [DT] + [HT] \} + 2[J=1 T_2]}{2[J=1 T_2]} T_{11} \quad (4)$$

The HT is a small impurity of about 1%. There is no special relation between the components, and they can change, because of the tritium radioactivity. Where the DT is enriched and the tritium minimized, Eq. 4 reduces to

$$T_1(\text{T in DT}) \approx \frac{3[DT]}{8[J=1 T_2]} T_{11} \quad (5)$$

The T_{11} values we need were measured in our laboratory in solid T_2 at about 6 K. The nmr frequency was 30 MHz, and we used the π - $\pi/2$ pulse sequence to measure T_1 . The $\pi/2$ -pulse length was 4-1/2 μ s. The receiver dead-time was 14 μ s, whereas the triton's transverse nuclear relaxation time was 22 to 38 μ s. The T_{11} values, listed in Table 1, are slightly different from the measured ones because of the correction for several percent HT and HD. The $J=1$ content is obtained by first measuring the $J=1$ time constant, τ , at various temperatures. These

results are shown in Fig. 1 for solid T_2 and D-T. We next calculate the % J=1 T_2 by the equation

$$[J=1 T_2] = \left\{ [J=1 T_2]_0 - [J=1 T_2]_\infty \right\} \exp(-t/\tau) + [J=1 T_2]_\infty \quad (6)$$

Here t is time and the quantities in brackets are mol %. The initial value (subscript "0") is determined from the sample history and the final value (subscript " ∞ ") from nuclear magnetic resonance.²⁵ For example, radiation keeps 2.3 % J=1 T_2 present even at 4 K, where the thermal J=1 T_2 concentration should be near-zero.

With a calculated % J=1 T_2 value in hand, we can calculate T_{11} . The triton T_{11} data shown in Table 1 is indeed comparable to that of the proton. This is further illustrated by the T_{11} plot of Fig. 2. The proton-in-HD data is that of Hardy and Gaines and Honig. There is considerable uncertainty at low J=1 values. The triton-in- T_2 data was taken at 5.3 and 7.0 K. Nevertheless, we use the theory described here as a framework for our D-T data, and it appears to fit.²⁶

We cannot use D_2 -DT- T_2 solutions to prove the validity of Eqs. 4 and 5. The reason is that we can measure only the triton and not the deuteron. Hence, we do not know how much J=1 D_2 exists, except at the start. Unfortunately, we need both the J=1 D_2 and J=1 T_2 concentrations to calculate T_{11} .

A problem should be mentioned here. The T_1 for protons in H_2 is constant from 4 to 10 K.²¹ At some J=1 H_2 concentrations, it stays constant even to 1.4 K, while at others, it drops by half. However, the time constant is not constant at all for protons in HD,²⁷ as shown in

Fig. 3. The HD shown there were Honig's two most impure samples with 0.02-0.07% J=1 H₂. Also shown in Fig. 3 is the triton in 95% molecular DT. It was held 18 hours at 8.3 K to allow de-excitation of the J=1 species. At 20 hours of age, we estimate 0.3 to 0.4% J=1 T₂. The DT data shows the same shape as that of the HD. In summary, T₁ is not a constant with temperature for HD and DT even though our theory implies that it is.

Experiments with D-T

We now use the theory of Eqs. 4 and 5 and apply it to actual D-T samples. We first write

$$Q = \frac{[DT] + [HT]}{[T_2]} \quad (7)$$

and obtain

$$\alpha = \frac{3QT_{11}}{8(T_1 - T_{11})} \sim \frac{3QT_{11}}{8T_1} \quad (8)$$

Q may be directly measured by collision-induced infrared spectroscopy.²⁸ The coefficient α is the fraction of T₂ in the J=1 state, which infrared spectroscopy cannot measure. The approximation T₁ >> T₁₁ is usually true.

We obtain T₁₁ by measurement of pure tritium. At 6 K, our data may be fit, for 2 ≤ $\alpha[T_2]$ ≤ 39% by

$$T_{11}(s) = 1.9483 \times 10^{-3} + 2.1244 \times 10^{-4} \alpha[T_2] + 4.2900 \times 10^{-4} \{\alpha[T_2]\}^2 - 7.4139 \times 10^{-6} \{\alpha[T_2]\}^3 \quad (9)$$

The first obvious experiment is to copy Honig and just wait. This result is shown in Fig. 4 for a T_2 -DT- D_2 sample brought from room temperature to the solid temperature of 6 K. The memory decreases, then rises somewhat, but we are worse off than we were. The reason in terms of Eq. 8 is this. The tritium radioactivity converts $J=1$ T_2 and D_2 to the $J=0$ form and α decreases. Eventually, this creates as much $J=1$ species as it destroys and α saturates. We have: $Q \sim 1.8$, $T_{11} \sim 2 \times 10^{-3}$ s, and $T_1 \sim 0.1$ s so that about 1% of the T_2 (and probably D_2 as well) is in the $J=1$ state. This is close to the T_{11} minimum, the worst possible place. In fact, we have probably just crossed to the low- $J=1$ side of the minimum. In any case, waiting does not work. The reason why may be seen from Fig. 1. We previously found that the $J=1 \rightarrow 0$ time constant in pure T_2 had a maximum rate at 10 K.^{25,29,30} We see that the same is true for solid 25 mol% T_2 -50 DT-25 D_2 . At 4 to 6 K, the time constant is 3 to 6 hours, which is too slow, because the $J=1$ T_2 is recreated by the tritium radioactivity with a probable 100-hour time constant.³¹

In order to test the polarization memory time theory, we decided to next add normal(n)- H_2 , which contains 75% $J=1$ H_2 at room temperature. In theory, Q should stay the same because we are not adding $J=1$ T_2 , but T_{11} should increase because we are increasing the total $J=1$ concentration. We first held T_2 -DT- D_2 at 9 to 11 K for four hours to reduce the $J=1$ concentration to a minimum. Then, we raised the temperature to 14.5 K and added a known amount of nH_2 as the liquid. We then heated to 21 K to liquefy everything and hoped that refluxing would cause good mixing. The results are shown in Table 2. First, the

calculated % of T_2 as $J=1$ is low, on the order of a few percent. We expected 2% from our earlier T_2 work. The $Q=1.86$ sample was remelted in order to see if better mixing would occur. Unfortunately, T_1 decreased instead, probably because of the constant loss of $J=1$ H_2 to the $J=0$ form. The 2.6-second T_1 value with a 4-minute mix appears to agree with the theory quite well.

We next turn to the enriched molecular DT samples, whose synthesis has already been described. The samples contained from 90 to 96 mol% DT (runs #5 to #8).³¹ Figure 5 shows the measured triton polarization memory times at three temperatures in the first hours after synthesis. The time T_1 increases because the $J=1$ T_2 is catalyzed away. We cannot explain why the 8.3 K sample does not change. The results are summarized in Table 3 as the zero-percent nH_2 data. We assume that the DT degrades toward about 50% and the T_2 toward 25% with a 100-hour time constant, and we then recalculate Q . Then we calculate α by simultaneously solving Eqs. 8 and 9, where we replace $\alpha[T_2]$ by $2\alpha[T_2]$ in Eq. 9. This assumes the same amount of $J=1$ D_2 is present at every moment with the $J=1$ T_2 . From Table 3, we see that the first measured percent of T_2 that is $J=1$, 100α , is 24% to 40%, where we expected 75% at the instant of synthesis. After several hours, this percent has fallen to 3% to 7% and T_1 has increased.

We then heated the DT samples, added the nH_2 and quickly refroze to 5.3 K. Table 3 lists the initial T_1 values of 3.5 to 7.4 s. Run #5 is also shown with a 25 T_2 -50 DT-25 D_2 sample, with both measured at 5.3 K (the latter sample is the same as in Fig. 4). Both decay with a

3-1/2 hour time constant, but the DT-nH₂ sample is always a factor of 20 higher. This is the largest increase we have been able to achieve by manipulating the D-T ingredients to date. Figure 6 has a logarithmic time scale to emphasize how polarization must be done: quickly.

The nH₂ contributes virtually all the J=1 species that determine T₁₁ in Eq. 9, so we may quickly solve Eq. 8. The results, listed in Table 3, say that the percent of T₂ in the J=1 state amounted to 12 to 25%, assuming perfect mixing. It appears that additional J=1 T₂ was formed during the heating and mixing sequence in addition to imperfect mixing. We have no reason at this point to doubt the theory, even though mixing results were disappointingly low.

We consider liquid DT for a moment. It is not possible to polarize it directly because: 1) 21 K is too hot to allow high polarization of even electrons with existing DC magnetic fields, and 2) most free hydrogen atoms have recombined and there are not enough pumps. There remains the possibility of polarizing at low temperature and then heating to the liquid. Two such DT-nH₂ results are shown in Fig. 7, where the samples were heated over a couple of minutes from 5.5 K into the liquid. The triton polarization memory time increases by about a factor of 3 to 4. The downward drift seen in Fig. 7 is caused by decay of the J=1 H₂.

To summarize, we have increased the triton polarization memory time 20-fold by manipulation of the hydrogen ingredients in ways suggested by the theory. We did not achieve T₁ values as large as we had hoped for, but we believe this is caused by imperfect liquid mixing and J=1 formation during heating. Our approach of looking for the most efficient self-catalysis of the J=1 to J=0 species has had only limited success.

Another explanation is assume that the paramagnetic mechanism has appeared to limit T_1 , i.e., that the hydrogen atoms are imposing the limit. We believe 10 s to be too short for this mechanism, and its appearance would be truly unfortunate. The mechanism could be removed by polarizing all the free electrons on the atoms, but pumping with microwaves to cause dynamic nuclear polarization would lower the electron polarization and bring the paramagnetic mechanism back.

Considerations for Future DT Purification

We clearly want the purest molecular DT we can get. An obvious way would be a distillation column. The following model was constructed from experience gained in the design of the purification system on the Tritium Systems Test Assembly at Los Alamos National Laboratory.³²

The design we use here is a single column run in the batch mode. The column is modeled to be 12.7 mm internal diameter and 2.03 m in height. The starting mixture of D-T is at the bottom of the column in the reboiler. The total charge of hydrogen amounts to 3.3 mol. A 12 W heater holds the column at about 24.5 K with a hydrogen vapor pressure of 105 kPa. The hydrogen isotopes are removed from the top of the column, with the most volatile hydrogens distilling off first. The model calculates the compositions of the hydrogens in each of 8 cuts, as they pass out of the top of the column.

Table 4 shows the calculated model results for two D-T compositions. The top section inputs regular D-T (25 mol% T_2 -50% DT-25% D_2). Cut #8 results in 97.4% DT, so that the single column does

not produce DT pure enough for us. The result is slightly better than the 96% obtained by our chemical synthesis.

The bottom section of Table 4 shows the results with a starting mixture out of our synthesizer: 95% molecular DT. The results represent the gas phase at the top of the column. Whether a number as small as 0.001% T_2 could actually exist at the end of a chilled outlet pipe is unknown.

We first calculate the polarization time constants for the "as-is" product emerging from the distillation column. The fraction of $J=1 D_2$ and $J=1 T_2$ is that of the thermal equilibrium at 24.5 K. We use the partition functions:³³

$$[J=1 D_2] = [D_2] \frac{3\exp(-86.0/T)}{2 + 3\exp(-86.0/T)} \quad (10)$$

and

$$[J=1 T_2] = [T_2] \frac{9\exp(-57.6/T)}{1 + 9\exp(-57.6/T)} \quad (11)$$

The $J=0$ -to- $J=1$ energies are 57.6 and 86.0 K for T_2 and D_2 , and T is the temperature. At 24.5 K, 46% of the T_2 and 4.3% of D_2 is in the $J=1$ state.

We assume that the distillation column output is instantaneously frozen to 4 K in the sample cell. We next use Eqs. 10 and 11 to calculate the triton memory times, which are listed Table 5 for DC magnetic fields of about 1 T. The memory time is calculated to be over 100 seconds (at 10 to 30 MHz nuclear magnetic resonance frequency), and it could be longer if the Hardy and Gaines' low $J=1$ data is correct.²⁴

However, the relaxation time could also be shortened by $J=1$ T_2 adsorbed in the lines on the way to the cell plus decomposition of the DT itself over the finite time of sample collection.

The single column system considered would probably cost a half-million dollars. It is not possible to borrow the continuous-flow systems now being used at LANL, Mound and Savannah River, even if they were available. This is because they use the cold output gas to cool the input gas, whereas our product must always be kept cold to avoid decomposition.

For what they cost, a distillation system does not do very well. This is because it does not address the real problem, which is to lower the $J=1$ species, especially the $J=1$ T_2 . What is really needed is a $J=1$ -to-0 converter - a machine that does not presently exist. Although such conversion is commonly used for H_2 in the liquid, the small $J=1$ -to-0 energy of 57.6 K for T_2 requires us to go to 12 K or below to do the conversion, as may be seen by calculating thermal $J=1$ percents from Eq. 10. The difficulty lies not in the catalysis. A monolayer of T_2 on a ferric oxide/aluminum oxide catalyst at 4 K will surely be converted swiftly to the $J=0$ state. The problem is to remove the strongly adsorbed gas without re-exciting the $J=1$ T_2 . Under less demanding circumstances, Cunningham used a heat pulse to drive adsorbed H_2 off alumina at 20.4 K.³⁴

It is easy to show that the output of a chemical synthesizer is adequate if only the $J=1$ -to-0 catalysis is carried out. Some calculated numbers are shown in Table 6. The DT is assumed to approach 50% and the T_2 25% with a 100-hour time constant. The purity of the mixture is

partly given by Q , as defined in Eq. 7. We see that the DT decomposition results in a constant lowering of Q . We now imagine a catalyst lowering the fraction of T_2 that is $J=1$, α . Suppose we adsorb a monolayer of 95% DT-1% T_2 on a weak catalyst like alumina. It may take hours to catalyze almost all T_2 to the $J=0$ state at 4.2 K, but almost none of the tritium beta particles should stop in the monolayer. We then blow off the DT with a heat pulse and collect it in the sample cell. A weak catalyst was used so that the heat pulse produces minimum $J=1$ T_2 . If only 5% of the T_2 is converted to the $J=1$ form, we calculate a triton memory time of 144 seconds. One hour later, it is 115 s. During this hour, nuclear polarization could be carried out.

Improving the synthesizer to make 99% DT rather than 95% is not a big help compared to somehow reducing the total T_2 . If we have 95% DT-0.1% T_2 with 5% of the T_2 in the $J=1$ state, then we expect a triton memory time of over a thousand seconds. The decomposition of the DT will decrease T_1 to 400 s after an hour. In any case, triton memory times of over a thousand seconds seem possible. This assumes that the mechanism remains one of spin-rotation, and that no paramagnetic mechanism appears.

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Figure Captions

- Fig. 1. The $J=1$ reaction time, τ , for T_2 in solid D-T and T_2 has a maximum rate at 10 K.
- Fig. 2. Inherent triton polarization memory time T_{11} as a function of $J=1$ concentration, showing the similar behavior of protons in HD and tritons in T_2 . The HD data was taken at 4.2 K and the T_2 data at about 6 K.
- Fig. 3. Comparison of the triton polarization memory time for protons in solid "impure" HD and tritons in one of the purest DT syntheses. The DT may contain 0.3 to 0.4% $J=1 T_2$.
- Fig. 4. Triton polarization memory time of 25% T_2 -50 DT-25% D_2 brought quickly from room temperature to the solid phase at 6 K.
- Fig. 5. Triton polarization memory time of four enriched molecular DT samples kept at 6 to 10 K. The reason for the differing behavior at 8.3 K is unknown.
- Fig. 6. Twenty-fold increase in the triton polarization memory time in going from regular T_2 -DT- D_2 and enriched DT containing nH_2 . The samples are solids at 5.3 to 6 K.
- Fig. 7. Increase of the triton polarization memory time in liquefying enriched DT- nH_2 samples.

Table 1. Summary of proton and triton longitudinal relaxation time data in solid H₂ and HD at 4.2 K and solid T₂ at about 6 K. The numbers in parentheses are powers of ten.

% J=1 Hydrogen	T ₁₁ (ms) H in H ₂ Meyer, 29 MHz	T ₁₁ (ms) H in H ₂ Gaines, 30 MHz	T ₁₁ (ms) H in HD Hardy & Gaines, 9 MHz	T ₁₁ (ms) H in HD Honig, 63 MHz	T ₁₁ (ms) T in T ₂ this work, 30 MHz	T ₁ (ms) H in HD Honig, 63 MHz
40	210	240	--	216	220	310
20	85	115	76	105	115	250
10	26	37	21	31	35	130
6	11	16	10	9	15	60
4	6	9	5.1	3.3	8	33
2	2.5	3.2	2.1	1.1	4*	22
1	--	1.7	1.1	0.7	--	25
0.6	0.8	0.9	0.8	0.5	--	30
0.4	0.9	0.9	0.8	0.4	--	38
0.2	--	--	1.1	0.5	--	100
0.1	--	--	1.5	1.6	--	600
0.04	--	--	6.6	4.3	--	4(3)
0.01	--	--	75	15	--	6(4)
4(-3)	--	--	--	16	--	2(5)
1(-3)	--	--	--	12	--	5(5)
4(-4)	--	--	--	11	--	1(6)
1(-4)	--	--	--	6	--	2(6)

* extrapolated from 2.4%

Table 2. Triton polarization memory time constants for two T₂-DT-D₂ mixtures at 5.3 K. The time is measured from the addition of nH₂ to the start of the T₁ measurement.

First Measured T ₁ (s)	Time (min)	Q	% J=1 H ₂ after adding	T ₁₁ (s)	Calcd. % J=1 T ₂ , 100%
0.9	48	1.76	17	0.093	7
1.5	82	1.86	17	0.093	4
1.7	65	1.86	17	0.093	4
1.8	48	1.86	17	0.093	4
1.8	33	1.86	17	0.093	4
2.1	18	1.86	17	0.093	3
2.6	4	1.86	17	0.093	2

Table 3. Triton polarization memory time constants for enriched DT and DT-nH₂ mixtures. DT data was taken at 8 to 10 K; the DT-nH₂ data was generally taken at 5.3 K.

DT Run	T ₁ (s)	Time Since synthesis (min)	Q	% J=1 H ₂	T ₁₁ (s)	Calcd % J=1, 100α
#5	0.15	2	38	0	3.0(-3)	28
	0.63	220	28	0	2.0(-3)	3
	6.5	245	27	17	0.093	15
#6	0.25	100	74	0	2.2e-3	24
	0.45	1470	14	0	-	<6
	7.4	1480	14	27	0.17	12
#7	0.20	4	78	0	2.4(-3)	36
	0.65	235	48	0	2.0(-3)	4
	3.5	240	48	10	0.040	19
#8	0.24	4	185	0	2.3e-3	>40
	0.80	185	78	0	2.0e-3	7
	6.0	200	74	12	0.054	25

Table 4. Model results for a 2-m high, batch-mode, single distillation column running at 24.5 K and 105 kPa, with the cuts are taken at the top of the column as the product emerges. The top section is for regular 25% T₂-50 DT-25 D₂, and the bottom is for enriched 95% molecular DT.

Cut Number	Time each Cut (min)	Mols each Cut	Instantaneous Mol%				
			HD	HT	D ₂	DT	T ₂
Start	--	--	0.5	0.5	25.0	48.9	25.1
1	16.7	0.045	15.9	6.6	66.8	9.8	0.7
2	3.3	0.089	9.0	6.6	82.7	1.6	0
3	20.0	0.54	0.5	1.2	87.8	10.5	0
4	8.3	0.22	0	0	61.1	38.8	0
5	8.3	0.22	0	0	35.8	64.2	0
6	8.3	0.22	0	0	13.6	86.2	0.2
7	8.3	0.22	0	0	3.5	96.0	0.6
8	0.8	0.22	0	0	0.6	97.4	1.9
Start	--	--	0.5	0.5	2.4	94.8	2.4
1	16.7	0.045	17.1	9.2	12.4	61.1	0.094
2	3.3	0.089	8.6	8.4	19.6	63.4	0.001
3	20.0	0.54	0.3	0.9	9.1	89.7	0.003
4	8.3	0.22	0	0.02	2.1	97.85	0.005
5	0.8	0.22	0	0	0.79	99.20	0.011
6	0.8	0.22	0	0	0.29	99.68	0.035
7	0.8	0.22	0	0	0.18	99.71	0.11
8	0.8	0.22	0	0	0.15	99.50	0.35

Table 5. Calculated polarization memory time constants in the instantly frozen 4 K output of distilled 95% molecular DT. These results belong with the lower half of Table 1. The nuclear magnetic resonance frequencies are about 10 to 30 MHz for the triton.

<u>Cut Number</u>	<u>Triton T_1(s)</u>
Start	0.14
1	1.6
2	190
3	50
4	150
5	170
6	70
7	10
8	0.8

Table 6. Calculated 4 K solid triton memory times for chemically-synthesized molecular DT with J=1-to-0 conversion.

<u>Instantaneous Output</u>	<u>Hours since Synthesis</u>	<u>Q</u>	<u>Fraction T2 in J=1, α</u>	<u>Calculated Triton T₁(s)</u>
95%DT 1% T ₂	0	96	0.2	36
	1	77	0.2	29
	4	48	0.2	18
	0	96	0.05	144
	1	77	0.05	115
	4	48	0.05	73
	0	96	0.01	718
	1	77	0.01	577
	4	48	0.01	363
95% DT 0.1% T ₂	0	957	0.2	359
	1	281	0.2	105
	4	90	0.2	34
	0	957	0.05	1436
	1	281	0.05	422
	4	90	0.05	135
	0	957	0.01	7178
	1	281	0.01	2108
	4	90	0.01	676

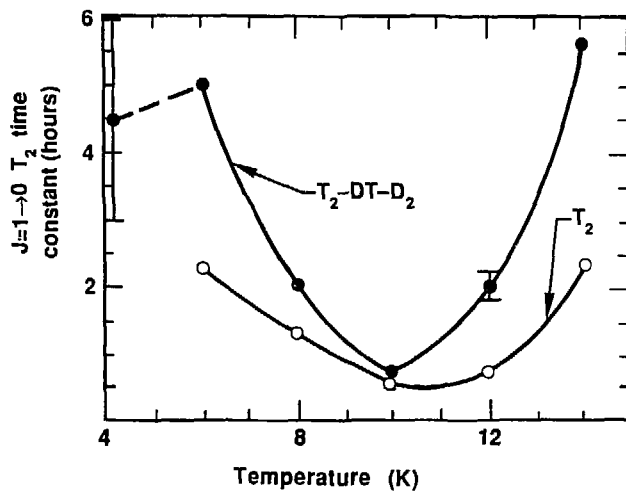


Fig. 1. The J=1 reaction time, τ , for T_2 in solid D-T and T_2 has a maximum rate at 10 K.

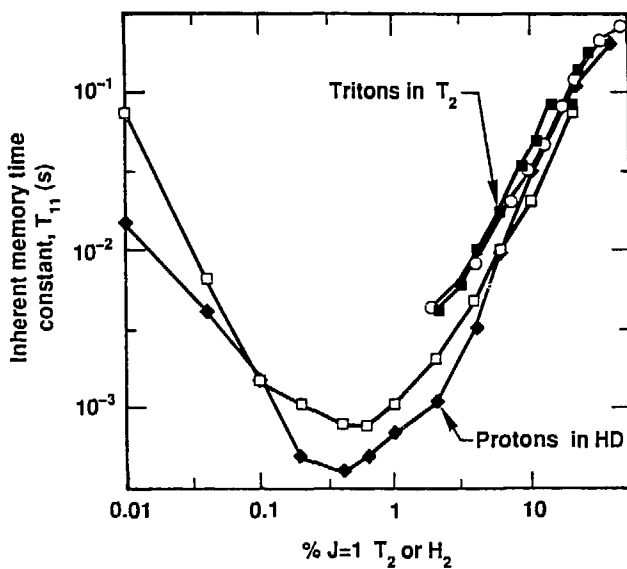


Fig. 2. Inherent triton polarization memory time T_{11} as a function of J=1 concentration, showing the similar behavior of protons in HD and tritons in T_2 . The HD data was taken at 4.2 K and the T_2 data at about 6 K.

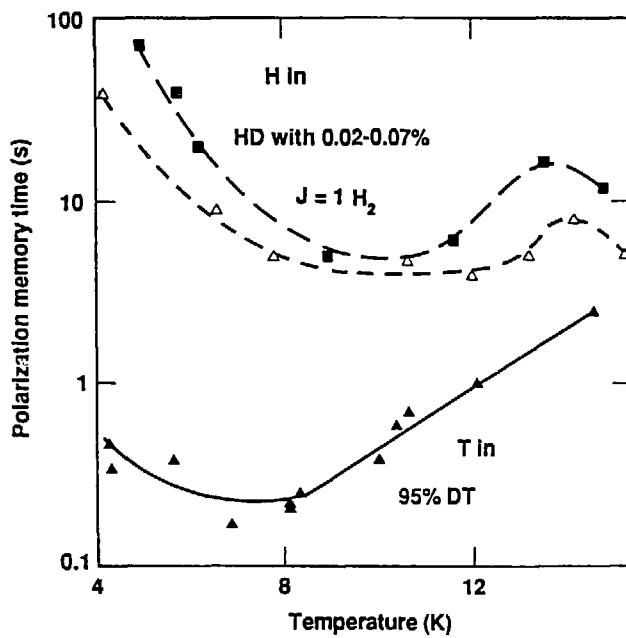


Fig. 3. Comparison of the triton polarization memory time for protons in solid "impure" HD and tritons in one of the purest DT syntheses. The DT may contain 0.3 to 0.4% $J=1 T_2$.

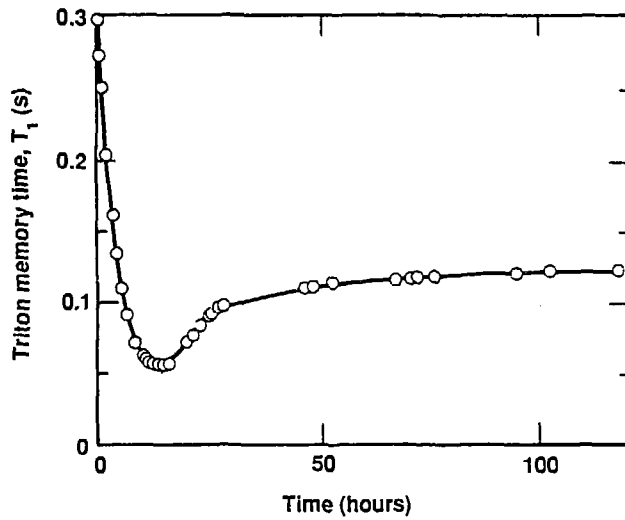


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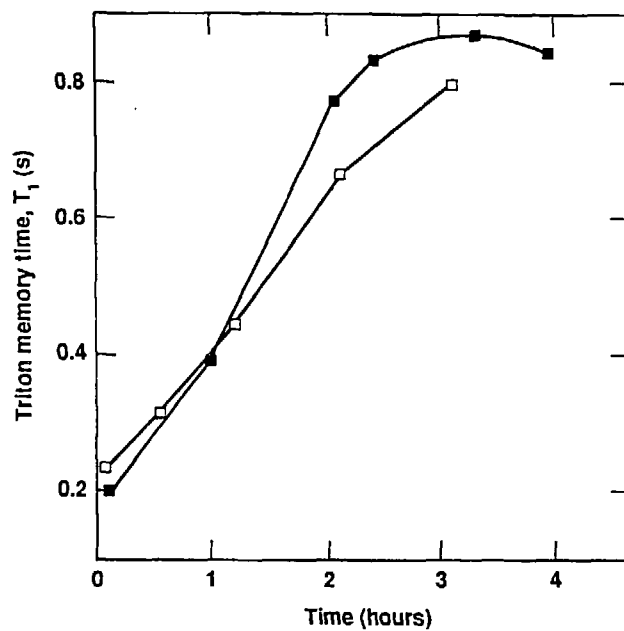


Fig. 5. Triton polarization memory time of four enriched molecular DT samples kept at 6 to 10 K. The reason for the differing behavior at 8.3 K is unknown.

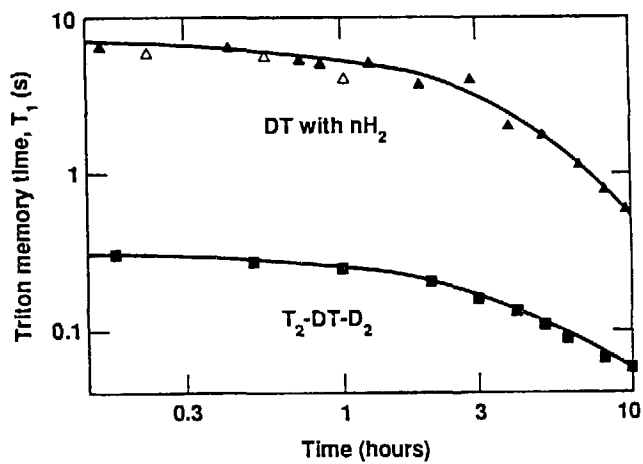


Fig. 6. Twenty-fold increase in the triton polarization memory time in going from regular T_2 -DT- D_2 and enriched DT containing nH_2 . The samples are solids at 5.3 to 6 K.

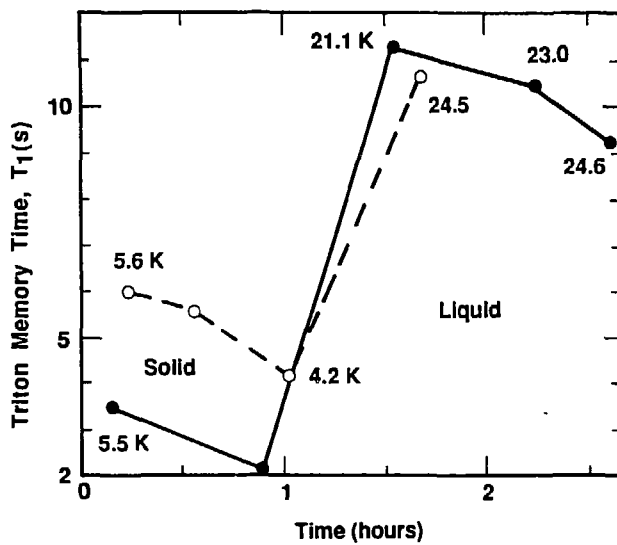


Fig. 7. Increase of the triton polarization memory time in liquefying enriched DT-nH₂ samples.