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NUCLEAR SPIN POLARIZATION
OF SOLID DEUTERIUM-TRITIUM

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ABSTRACT

MASTER

It appears that parallel alignment of deuteron and triton magnetic moments increases the cross section of the nuclear reaction $T(d,n) He^4$ by 50%, thereby promising a laser driver of perhaps half the original energy. Both "brute-force" and dynamic nuclear polarization are considered, and the many potential problems of the latter are considered. High nuclear polarization by the dynamic technique requires a small nucleus-to-unpaired electron ratio, a long longitudinal nuclear relaxation time and a short longitudinal electron relaxation time. Normal D-T is shown to be inadequate, and enriched and possibly very pure molecular DT will be required. The key variable is the nuclear relaxation time, which can either depend on the interaction with rotationally excited impurity molecules or on paramagnetic defects formed by the tritium radiation. Radiation-induced DT decomposition and rotational catalysis will combat one another to affect the DT purity. The expected atom density and fractionation effects are considered. There exists one frequency at which both D and T atoms can be pumped.

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INTRODUCTION

An exciting new idea by Goldhaber has entered the hydrogen fusion picture.¹ He suggested that the reaction cross section for deuterium-tritium (D-T) may be increased by polarizing the nuclear magnetic moments. The triton has a nuclear spin of $\frac{1}{2}$ and so splits into two magnetic sublevels (quantum number

$m_I = \frac{1}{2}$ and $-\frac{1}{2}$). The deuteron has a nuclear spin of 1 and has three sublevels ($m_I = 1, 0$ and -1). If almost all of the (D,T) nuclei are in either the $(1, \frac{1}{2})$ or the $(-1, -\frac{1}{2})$ sublevels, i.e., both with parallel spins, the nuclei are "polarized" for the purposes of fusion.

Suppose a random mix of nuclear spins has a relative cross section of 1. So do the polarized $(0, \frac{1}{2})$ or $(0, -\frac{1}{2})$ combinations. Theory shows that the anti-parallel $(1, -\frac{1}{2})$ and $(-1, \frac{1}{2})$ arrangements have a relative cross section of $\frac{1}{2}$. The parallel combinations mentioned above have a cross section of $3/2$, 50% higher than the random number. This 50% improvement in cross section could allow a laser of perhaps half the originally-contemplated size to be used.² This could result in a very large savings of money in the laser construction. The final inducement is a calculation done at the Lawrence Livermore National Laboratory by More, who found that the laser shot should not destroy the nuclear polarization before the nuclear reaction occurs.³

How to actually polarize D-T is entirely another matter. There are two possible avenues. One is being pursued by Honig at Syracuse. His approach is to cool solid hydrogen to a few degrees milliKelvin and "brute-force" magnetically align the nuclear moments.⁴ A variation of this method is to lower the magnetic field until the Larmor frequency envelopes of the nuclear moments overlap.⁵ Then one kind of nucleus can transfer its polarization to another kind. Honig has perfected his method on HD and is presently working on D₂. D-T, however, poses a problem because of the 200 mW/g radioactive

self-heating. Honig proposes to try to polarize thin layers of D-T on solid blocks of HD.

Dynamic Nuclear Polarization

The approach we shall consider here is dynamic nuclear polarization, which is used for most high energy nuclear spin-polarized targets. This method requires "pumpers" in the form of free atoms with electron magnetic moments. Such atoms are formed constantly by radiation damage in solid D-T. A DC magnetic field is used to align the electron moments. Because the electron moment is a thousand times larger than the nuclear moments, we need cool only to 1 to 4 K rather than to milli-Kelvin. This removes an enormous constraint, given the tritium heat.

Next, we use microwave radiation to pump the levels shown in Figs. 1 and 2. Figure 1 shows the sub-levels for the interacting system of the spin- $\frac{1}{2}$ electron and the spin- $\frac{1}{2}$ triton. The spin quantum numbers, m_S and m_I , refer to the electron and nucleus. The interaction is by weak dipolar coupling of the magnetic moments. The transitions (m_S, m_I) of $(\frac{1}{2}, -\frac{1}{2}) \rightarrow (-\frac{1}{2}, -\frac{1}{2})$ and $(\frac{1}{2}, \frac{1}{2}) \rightarrow (-\frac{1}{2}, \frac{1}{2})$, where $\Delta m_I = 0$, are electron spin resonance (e.s.r.) transitions. These are labeled in Fig. 1 by the longitudinal (loss-of-energy) relaxation time T_{1e} . Also shown in Fig. 1 are the nuclear magnetic resonance (n.m.r.) transitions, which are described by the longitudinal relaxation time. These transitions have $\Delta m_S = 0$. We note that the thermal energy at 4 K is large compared to splitting between the

two lowest energy levels. At equilibrium, then, the $(-\frac{1}{2}, \frac{1}{2})$ and $(\frac{1}{2}, -\frac{1}{2})$ states are almost equally populated. However, if we use a 8 to 10 T magnet, this creates an energy difference between the $m_S = -\frac{1}{2}$ and $\frac{1}{2}$ states large compared to the thermal energy. All electron states will have $m_S = -\frac{1}{2}$, i.e., the electrons will be almost completely polarized by the DC magnetic field.

Now, we saturate the transition shown by the double arrow in Fig. 1. This is $(-\frac{1}{2}, -\frac{1}{2}) \leftrightarrow (\frac{1}{2}, \frac{1}{2})$ and it corresponds to a combined e.s.r.-n.m.r. transition. We could use the other transition $(-\frac{1}{2}, \frac{1}{2}) \leftrightarrow (\frac{1}{2}, -\frac{1}{2})$ if we want. By saturating the indicated transition, we equalize the populations of the two states involved. These decay by T_{1e} to lower states. We see that all paths lead to the very lowest state $(-\frac{1}{2}, \frac{1}{2})$. When this state fills up, the sample has a positive (because of $m_I = \frac{1}{2}$) nuclear polarization. We see that we can only polarize if T_{1e} is relatively short and T_{1n} is long. If the $(-\frac{1}{2}, \frac{1}{2})$ nuclei thermalize back into the $(-\frac{1}{2}, -\frac{1}{2})$ state because T_{1n} is short, we cannot hold our polarization. The rate equations for the spin $\frac{1}{2}$ - $\frac{1}{2}$ case produce this approximate nuclear polarization, P_n :^{6,*}

* The polarization for the triton is defined as

$$P_n = \frac{\left[\begin{matrix} -\frac{1}{2} & \frac{1}{2} \end{matrix} \right] + \left[\begin{matrix} \frac{1}{2} & \frac{1}{2} \end{matrix} \right] - \left[\begin{matrix} -\frac{1}{2} & -\frac{1}{2} \end{matrix} \right] - \left[\begin{matrix} \frac{1}{2} & -\frac{1}{2} \end{matrix} \right]}{\left[\begin{matrix} -\frac{1}{2} & \frac{1}{2} \end{matrix} \right] + \left[\begin{matrix} \frac{1}{2} & \frac{1}{2} \end{matrix} \right] + \left[\begin{matrix} -\frac{1}{2} & -\frac{1}{2} \end{matrix} \right] + \left[\begin{matrix} \frac{1}{2} & -\frac{1}{2} \end{matrix} \right]}$$

where the brackets contain the populations of each state, $[m_S, m_I]$. The polarization is the relative difference between the positive and negative nuclear states. The same is true for the deuteron, except that the sum of states in the denominator includes the two spin-0 states.

$$P_n = \frac{-\beta P_0}{\beta(f+1) + 2f} \quad (1)$$

Here, P_0 is the initial electron polarization, which should be close to -1 with a 8 T magnet. The quantity β describes the level of microwave power used to pump the transition, and

$$f = \frac{N_n T_{1e}}{N_e T_{1n}} \quad (2)$$

Where, N_n is the number of nuclei and N_e the number of hydrogen atoms pumping them. To get P_n as near 1 as possible, we want a large β and a small f . To obtain this last requirement, we want T_{1e} small, T_{1n} large (as we noted above) and as many atoms-per-nuclei as possible. Equation 2 is the centerpiece of all starting work for dynamic nuclear polarization.

Figure 2 shows the same behavior for the electron and deuteron with weak dipolar coupling between them. The saturation microwave frequency saturates two sets of levels at once. The nuclei will again cascade to the bottom by T_{1e} relaxation. If T_{1n} is long, the $(-\frac{1}{2}, 1)$ state will fill up and the deuterons will be positively polarized. Equation 2 holds also in this case.

Dynamic nuclear polarization has not yet been done. The closest case is that of Solem with solid HD at 1.3 to 4 K and 1.2 to 1.3 T.^{7,8} Solem obtained a proton polarization of 3/4% and a deuteron polarization of 0.3 to 0.4%. He put oxygen into the HD to lower T_{1e} to about 1 ms. The T_{1n} 's were 30 to 100 s. Radiation provided the atoms, and he guessed that

N_n/N_e was about 10^4 , although it could have easily been less. The best numbers give an f value of 0.1, small enough to have produced better polarization than what he saw.

Forms of D-T

A problem with D-T is its form. The equilibrium 50 D-50 T room temperature mixture, if cooled to cryogenic temperatures, really has in it:

$$\begin{array}{lcl}
 & & \left\{ \begin{array}{l} 3\% J=0, I=0 D_2 \\ 14\% J=0, I=2 D_2 \\ 8\% J=1 D_2 \end{array} \right. \\
 25 \text{ mol } \% D_2 & & \\
 50\% DT & & \\
 25\% T_2 & & \left\{ \begin{array}{l} 6\% J=0 T_2 \\ 19\% J=1 T_2 \end{array} \right. \quad (3)
 \end{array}$$

Here, J is the molecular rotational quantum number and I is the total nuclear spin quantum number. In the above mixture, we cannot polarize the 9% of $J=0$ species, because they have no magnetic moment to work with. Also, we have spin-1 tritium and spin-2 deuterium - both more complicated than our simple models of Figs. 1 and 2. The simple models are realized in the 50% molecular DT, in which the triton and deuteron at magnetic fields greater than 1 T, are effectively decoupled from one another.

We are not through. The tritium beta particle makes a fine catalyst for $J=1 \rightarrow 0$ de-excitation.⁹⁻¹¹ At 22 K, the equilibrium mixture, formed after about a day, is:

25 mol% D_2

{ 4% $J=0, I=0 D_2$
21% $J=0, I=2 D_2$

50% DT

25% $J=0 T_2$

(4)

Now we have 29% of the sample with no moment to polarize. This includes a full third of the tritium.

It is clear that we have to go to pure molecular DT in order to get an all-polarizable material and that meets the simplest spin models. We have made 90% DT in our laboratory by the reaction of lithium tritide and deuterated methanol. A faster but more capital-intensive means is to use cryogenic distillation.

Nuclear Magnetic Relaxation

The first big question centers on the longitudinal nuclear magnetic relaxation time, T_{1n} . Figure 3 shows this time for protons and deuterons in HD doped with $J=1 H_2$ "impurities."¹²⁻¹⁶ The deuteron T_{1n} is longer than that of the proton, because the deuteron magnetic moment is smaller, and it interacts less strongly with its surroundings. If we ignore radiation damage, we would expect the corresponding graph for DT to look similar to Fig. 3. This is because both the proton and triton have a nuclear spin of $\frac{1}{2}$ and very similar n.m.r. frequencies (42.6 and 45.4 MHz at 1 T). Our first triton T_{1n} measurements with large $J=1$ compositions indeed are similar to the corresponding proton values.

The presence of the T_{1n} minimum in Fig. 1 is disappointing indeed. The $J=1$ H_2 molecule has a molecular quadrupole moment, which interacts with the molecular rotation to split the rotational J -levels into a band of narrowly spaced sub-levels. When the n.m.r. energy level difference is the same as these sub-levels, energy flows from the nuclear magnetic to the rotational system.^{17*}

Unfortunately, the radiation damage in D-T (1.072×10^{15} Bq/mol T) completely confuses the issue. First, the atoms and ions formed by radiation catalyze the $J=1 \rightarrow 0$ de-excitation with time constants τ_J^T for the triton and τ_J^D for the deuteron. The time constant in pure tritium decreases as the temperature falls, because more paramagnetic defects build up.¹¹ At 4.2 K, we might expect τ_J^T to be minutes. We might think that τ_J^D would be longer because of the smaller magnetic moment of the deuteron.

The other reaction that is going on is the radiation-catalyzed decomposition

where we presume that the high temperature equilibrium amounts of $J=1$ hydrogen are formed ($1/3 D_2$; $3/4 T_2$). We do not know what the true $J=1$ equilibrium is, but we do see a $J=1$ infrared

* T_{1n} is usually called a spin-lattice relaxation time, but that is not correct here. For unirradiated solid hydrogen, T_{1n} is a spin-rotation time. The rotation-lattice interaction time is much faster.

T_2 peak that shows that this species is being constantly created even at low temperatures. Equation 5 creates $J=1$ species with a time constant τ_{ex} , which is of the order of 100 to 140 hours.¹⁸ If the fraction of either $J=1$ T_2 or $J=1$ D_2 is $c(l)$, then the two mechanisms compete in the reaction:

$$\frac{dc(l)}{dt} = \frac{c(l)_{\infty} - c(l)}{\tau_{ex}} - \frac{c(l)}{\tau_J} = \frac{c(l)_{\infty}}{\tau_{ex}} - \frac{c(l)}{\tau_J} \quad (6)$$

The approximation is valid if $\tau_{ex} \gg \tau_J$. The solution is:

$$c(l) = c(l)_e [1 - \exp(-t/\tau_J)] + c(l)_0 \exp\left(-\frac{t}{\tau_J}\right) \quad (7)$$

The equilibrium $J=1$ concentration at long time is

$$c(l)_e = \frac{\tau_J}{\tau_{ex}} c(l)_{\infty} \quad (8)$$

The quantity $c(l)_{\infty}$ is the value that the $J=1$ composition tries to reach, i.e., the presumed high temperature value, and $c(l)_0$ is the initial concentration. For an equimolar mix, $c(l)_{\infty}$ is 0.188 for the tritium and 0.0833 for the deuterium, if a high temperature equilibrium is attained. Suppose we wanted $c(l)_e$ to be 10^{-4} . Then Eq. 8 predicts that we need $J=1 \rightarrow 0$ time constants of 4 minutes for tritium and 9 minutes for deuterium. We do not know whether the paramagnetic catalysis in the solid can be this fast. If the $J=1$ equilibrium is not a high temperature one, then $c(l)_0$ will be smaller and τ_J can be longer.

We are not yet finished. The paramagnetic defects in the solid can also lower T_{1n} .^{4,8} It is possible that so many defects are present that they control T_{1n} . The defects could create a relaxation time $(T_{1n})_p$ which combines with the old quadrupolar time, T_{1n} , to give

$$\left(\frac{1}{T_{1n}}\right)_{\text{total}} = \frac{1}{T_{1n}} + \left(\frac{1}{T_{1n}}\right)_p \quad (9)$$

The dotted line in Fig. 3 shows the effect of a $(T_{1n})_p$ value of 20 ms. The dip is still evident, but, otherwise, T_{1n} is not a function of the J=1 concentration.

We are presently initiating experimental work to find which mechanism, quadrupolar or paramagnetic, determines the triton T_{1n} in solid D-T. If it is the former, then the purity of the enriched DT will be critical. There will be a possibility of achieving a long T_{1n} , although probably not as long as that seen for HD. On the other hand, the crucial Eq. 2 relationship may not hold. We may have to dope with oxygen to lower T_{1e} , which, in turn, could lower T_{1n} . If the paramagnetic mechanism dominates, then the purity of the DT is no longer a crucial issue. T_{1n} will be short, although it appears that the Eq. 2 condition will automatically be met.¹⁹ This is because the effect of a paramagnetic defect on a large electron moment is expected to be greater than its effect on a smaller nuclear moment. In this case, we would expect T_{1e} to be microseconds. However, the small T_{1n} now makes saturation of the microwave transition an almost explosively short event.

Whether it would be possible to polarize so quickly remains to be seen. The protons in solid ammonia have been polarized in minutes.^{20,21} How much faster it can be done is unknown.

Atoms in the Solid

In the gas phase, at 1.1 to 28 kPa and 295 to 373 K, five hydrogen atoms are formed for every ion pair created by the tritium beta particle.²² These atoms most likely come from these sources.^{23,24}

directly formed from beta particle	1 atom	
$H_2^+ + H_2 + H_3^+ + H$	1	
$H_3^+ + e^- + 3H$	<u>3</u>	
Total	5	(10)

These reactions quite likely happen in the solid. In any case, atoms are seen by electron spin resonance in irradiated solid hydrogen.²⁵ No data is available for solid D-T and we must turn to H_2 instead. If the solid is irradiated with a beam, which is turned off with some atom density N_1 , then the decay of this density goes as

$$\frac{dN}{dt} = -kN^2 \quad (11)$$

because two atoms form one molecule. The recombination rate constant is k . The solution is

$$\frac{N}{N_i} = \frac{1}{1 + N_i k t} \quad (12)$$

and the 1/e time τ is

$$\tau = \frac{1.717}{k N_i} \quad (13)$$

Fragments of H_2 data have been taken from 1.4 to 5.4 K and 6.75 to 8.05 K.²⁶⁻²⁸ We may piece these together using Eqs. 12 and 13 to obtain these estimates of the atom density in solid H_2 .

Temp. (K)	Nucleus-to-atom ratio, N_n/n_e
8.05	2.5×10^5
6.75	2.0×10^4
4.2	8.3×10^3
1.4	3.8×10^2

(14)

The 1.4 and 4.2 K values are dubious, but they offer promise that large atom densities may be possible - one of the key ingredients in Eq. 2.

Another problem arises as to the type of hydrogen. We might expect 50 D - 50 T in an equimolar D-T mixture. However, considerable fractionation is seen in H_2 - D_2 mixtures, as illustrated in Fig. 4.^{29,30} We see that the lighter atom predominates over the heavier one. This is attributed to quantum tunneling, in which a cryogenic thermalized atom can jump through the potential barrier and exchange with a molecule.³¹⁻³³

The decrease in molecular vibrational zero-point energy tilts the exchange towards the formation of heavier hydrogen molecules and lighter hydrogen atoms. This effect occurs only in cold solids where the reactants are initially close to each other. Its importance for us is that we will now expect a preponderance of deuterium atoms, and we should expect to pump at the D atom frequency. It is conceivable that only H atoms could appear from the 1% impurity usually present in D-T.

Microwave Frequencies

We have seen that the pumping frequency for dynamic nuclear polarization is a double e.s.r.-n.m.r. transition. The basic frequency is the free electron frequency of $28.025B_0$ GHz, where B_0 is the external DC magnetic field in T. Next we add the hyperfine splitting, which comes from the interaction of the electron and nucleus on the atom. The triton, with spin $\frac{1}{2}$, splits into a doublet and the deuteron, with spin 1, splits into a triplet. It is easy, then, to tell in e.s.r. which atom is which. But the nuclei we are interested in for polarization are in molecules next to the atoms. The weak dipolar coupling then adds sidebands to each line. We have these frequencies, ν .

$$\nu(T) = (28.025B_0 \pm 0.758) \pm 0.0454B_0 \quad (15)$$

for a total of 4 lines, and

$$\nu(D) = (28.025B_0; 28.025B_0 \pm 0.218) \pm 0.0065B_0 \quad (15)$$

for a total of 6 lines.

The complete energy level scheme is more complicated than what we showed in Figs. 1 and 2. The levels for a tritium atom coupled to a triton in a DT molecule is shown in Fig. 5. Three other schemes exist as permutations of D and T. In Fig. 5, m_1^* refers to the spin quantum number of the nucleus on the tritium atom. The usual relaxation processes are shown by T_{1e} and T_{1n} . We also have the n.m.r. relaxation time T_{1n}^* , which refers to relaxation only of the atomic nucleus. This time has been measured for H in H_2 and it is 3000 seconds.³⁴ This may be compared with a T_{1n} in normal H_2 of about 300 ms.³⁵ Also measured with T_{1n}^* was a T_{1e} of 50 ms.³⁴ Because $T_{1n}^* \gg T_{1n}, T_{1e}$, we need not consider the nuclear relaxation within the atom. We need only consider half of the diagram of Fig. 5, and this corresponds exactly to Fig. 1. With a one-frequency source, we can pump half of the tritium atoms or one-third of the deuterium atoms. Thus, N_n/N_e in Eq. 2 must be modified by these fractions. There exists a magnetic field B_0^* at which one of the inner triton lines will just match one of the deuteron lines. The lowest magnetic field that this overlap occurs at is:

$$-0.758 + 0.0454B_0^* = -0.218 + 0.0065B_0^*$$

$$B_0^* = 13.9 \text{ T}$$

(17)

This is a high but achievable magnetic field. We clearly will need a pumping source with a narrow enough bandwidth to just hit the line we want. If we cover several lines, we can pump nuclei into both positive and negative states and lose all chance at polarization.

Finally, we note the role of spin diffusion in the polarization process. Each atom pumps many nuclei, one after another. We are counting on the polarization, once formed on a neighboring molecule to wander away from the atom by spin diffusion. This leaves behind fresh molecules to be lined up. A calculated spin diffusion coefficient for protons in HD of $3 \times 10^{-16} \text{ m}^2/\text{s}$ would be plenty fast enough,³⁶ but data is lacking.

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References

1. R. M. Kulsrud, H. P. Furth, E. J. Valeo and M. Goldhaber, Princeton Plasma Physics Laboratory Report PPPL-1912 (June, 1982).
2. Y. L. Pan and S. T. Hatchett II, Lawrence Livermore National Laboratory, Livermore, California 94550, private communication, 1985.
3. R. M. More, Phys. Rev. Lett. 51, 396 (1983).
4. A. Honig, Syracuse University, Syracuse, New York 13210, private communication, 1985.
5. A. Honig and H. Mano, Phys. Rev. B 14, 1858 (1976).
6. C. D. Jeffries, Dynamic Nuclear Orientation (Interscience, New York, 1963), pp. 121-133.
7. J. C. Solem and G. A. Rebka, Jr., Phys. Rev. Lett. 21, 19 (1968).
8. J. C. Solem, Nucl. Instr. Methods 117, 477 (1974).
9. E. W. Albers, P. Harteck and R. R. Reeves, J. Amer. Chem. Soc. 86, 204 (1964).
10. R. Frauenfelder, F. Heinrich and B. Olin, Helv. Phys. Acta 38, 279 (1965).
11. J. R. Gaines, R. T. Tsugawa and P. C. Souers, Phys. Rev. Lett. 42, 1717 (1979).
12. W. N. Hardy and J. R. Gaines, Phys. Rev. Lett. 17, 1278 (1966).
13. F. Weinhaus and H. Meyer, Phys. Rev. 7E, 2974 (1973).

14. R. S. Rubins, A. Feldman, and A. Honig, Phys. Rev. 169, 299 (1968).
15. R. B. Bernardi, Proton Spin-Lattice Relaxation in Solid Hydrogen Deuteride over the Temperature Range 0.44 to 4.2°K, PhD Thesis, Syracuse University, Syracuse, NY (1972); University Microfilms, Ann Arbor, MI 48106, No. 73-7704. The data has been adjusted to 0.15 T by A. Honig and H. Mano of Syracuse University. Arnold Honig was the thesis supervisor.
16. A. Honig, Syracuse University, NY, 13210, private communication, 1983.
17. J. R. Gaines and P. C. Souers, "The Spin-Lattice Relaxation Time (T_1) in mixtures of Hydrogen Isotopes," in Advances in Magnetic Resonance, J. S. Waugh, ed. (Academic Press, Orlando, FL, 1985), Vol. 12, to be published.
18. P. C. Souers, E. M. Fearon and R. T. Tsugawa, J. Phys. D: Appl. Phys. 15, 1535 (1982).
19. A. Abragam and M. Goldman, Rep. Progr. Phys. 41, 395 (1978). See pp. 400, 414-415.
20. M. L. Seely, A. Amittay, M. R. Bergström, S. K. Dhawan, V. W. Hughes, R. F. Oppenheim, K. P. Schüller, P. A. Souder, K. Kondo, S. Miyashita, K. Morimoto, S. J. St. Lorant, Y-N Guo, and A. Winnacker, Nucl. Instr. Methods 201, 303 (1982).
21. W. Meyer, K. H. Althoff, W. Havenith, H. Riechert, E. Schilling and G. Sternal, Nucl. Instr. Methods 215, 65 (1983).

22. W. M. Jones and D. F. Dever, J. Chem. Phys. 60, 2900 (1974).
23. P. C. Souers, Hydrogen Properties for Fusion Energy,
(Berkeley, University of California Press, to be published
1986), Chapter 19.
24. J. B. A. Mitchell, J. L. Forand, C. T. Ng, D. P. Levac, R.
E. Mitchell, P. M. Mul, W. Claeys, A. Sen and J. Wm.
McGowan, Phys. Rev. Lett. 51, 885 (1983).
25. M. Sharnoff and R. V. Pound, Phys. Rev. 132, 1003 (1963).
26. R. K. Leach, A Paramagnetic Resonance Study of Hydrogen Atom
Production and Recombination in Solid H₂ from 1.4 to 8 K,
thesis, University of Wisconsin, Madison, Wisconsin (1972),
University Microfilms, Ann Arbor, Michigan 48106, 1972, No.
72-23060.
27. A. Y. Katunin, I. I. Lukashevich, S. T. Orozkamatov, V. V.
Sklyarevskii, Y. V. Suraev, V. V. Filippov, N. I. Filippov
and V. A. Shevtsov, Soviet Phys. - JETP Lett. 34, 357 (1981).
28. A. V. Ivliev, A. Y. Katunin, I. I. Lukashevich, V. V.
Sklyarevskii, V. V. Suraev, V. V. Filippov, N. I. Filippov
and V. A. Shevtsov, Soviet Phys. - JETP Lett. 36, 472 (1982).
29. H. Tsuruta, T. Miyazaki, K. Fueki and N. Azuma, J. Phys.
Chem. 87, 5422 (1983).
30. A. V. Ivliev, A. S. Iskovskikh, A. Ya. Katunin, I. I.
Lukashevich, V. V. Sklyarevskii, V. V. Suraev, V. V.
Filippov, N. I. Filippov and V. A. Shevtsov, Soviet Phys. -
JETP Lett. 38, 379 (1983).

31. R. J. LeRoy, E. D. Sprague and F. Williams, J. Phys. Chem. 76 546 (1972).
32. W. Siebrand, T. A. Wildman, and M. Z. Zgierski, J. Amer. Chem. Soc. 106, 4083 (1984).
33. W. R. McKinnon and C. M. Hurd, J. Phys. Chem. 87 1283 (1983).
34. A. Ya. Katunin, I. I. Lukashevich, S. T. Orosamatov, V. V. Sklyarevskii, V. V. Suraev, V. V. Filippov, N. I. Filippov, and V. A. Shevtsov, Phys. Lett. 87A, 483 (1982).
35. F. Weinhaus and H. Meyer, Phys. Rev. B 7, 2974 (1973).
36. T. Nakamura and M. Fujio, Progr. Theor. Phys. 54, 601 (1975).

Figure Captions

Fig. 1. Schematic of the magnetic levels of an electron and a triton with weak dipolar coupling.

Fig. 2. Schematic of the magnetic levels of an electron and a deuteron with weak dipolar coupling.

Fig. 3. Longitudinal nuclear relaxation times T_{1n} for protons and deuterons in solid HD with $J=1$ H_2 H_2 .

Fig. 4. Atomic fractionation seen in solid H_2 - D_2 .

Fig. 5. Weak dipolar coupling to a T nucleus in a nearby molecule by the electron in a T atom. The low-field hyperfine line is being pumped.

Fig. 1. Schematic of the magnetic levels of an electron and a triton with weak dipolar coupling.

Fig. 2. Schematic of the magnetic levels of an electron and a deuteron with weak dipolar coupling.

Fig. 3. Longitudinal nuclear relaxation times T_{1n} for protons and deuterons in solid HD with $J=1$ H_2 H_2 .

Fig. 4. Atomic fractionation seen in solid H₂-D₂.

Fig. 5. Weak dipolar coupling to a T nucleus in a nearby molecule by the electron in a T atom. The low-field hyperfine line is being pumped.