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THE RECOMBINATION OF ATOMIC HYDROGEN BELOW 1K

Saps Buchman*, Yueming Xiao*, Lois Pollak#, Daniel Kleppner#, and Thomas J. Greytak#

*GP-B Program, Hansen Laboratories of Physics, Stanford University, Stanford, CA 94305 U.S.A.
#Department of Physics, Massachusetts Institute of Technology Cambridge, MA 02139 U.S.A.

We have conducted an NMR study of the hydrogen recombination process in the temperature range 0.2K - 0.6K. Starting with a gas of doubly polarized atomic hydrogen we produce molecular ortho hydrogen. The nuclear spin polarization is quickly lost after the recombination process. The time constant for the de-polarization is less than 0.1s, and the initial nuclear spin temperature of the solid is larger than 4K.

The recombination of atomic hydrogen is one of the few chemical reactions simple enough to allow calculations from first principles. It is therefore highly desirable to study this fundamental process under well controlled conditions, so that the experimental results can be compared in detail with the theoretical predictions.

The experimental method used for this study is as follows. We first stabilize a gas of atomic hydrogen by polarizing the electron spins at a temperature of 0.2K to 0.6K in a magnetic field of 6.7T. Stable nuclear polarization follows automatically, with polarization times of the order of 1000 seconds (1,2). Figure 1 shows the hyperfine energy levels of a hydrogen atom in a magnetic field. It is the *b* hyperfine state which is stable and referred to as doubly spin polarized hydrogen. The recombination is then induced by flipping the electron spin. The techniques we use to produce and confine the doubly spin polarized hydrogen are described in our earlier work (2).

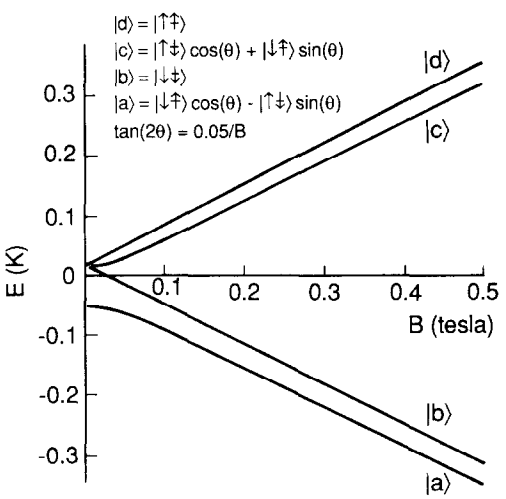

FIGURE 1
The hyperfine diagram of atomic hydrogen in a magnetic field. The plain and crossed arrows indicate the polarization of the electron and the proton spin.

The typical polarized atomic gas densities achieved in this experiment are of the order of 10^{16}atoms/cc at 0.3K. At these densities the dominant recombination process is the two-body surface recombination (3), with molecules forming close to the highest energy bound state; vibrational quantum number V = 14 and rotational quantum number J = 1 or 3 (4,5). We choose the *b+c* recombination channel since it has the following advantages: 1) The recombination rate is easily controlled by the *b* to *c* conversion rate. 2) The para hydrogen production is negligible. 3) Molecules form in a state with nuclear spin quantum numbers I = 1, M_I = -1. Note that the nuclear polarization has the opposite sign from the thermal nuclear polarization, a characteristic easily detectable by NMR. The *c* atoms are produced by ESR from *b* atoms at a frequency of 187GHz, for the 6.7T field.

Figure 2 is a diagram of our experimental cell. The cell contains a polarization chamber for the atomic gas, and an NMR chamber, where the stimulated recombination into molecular hydrogen takes place. The two chambers are connected by a 10mm length of 0.2mm diameter tube. By changing the helium level in the cell the chambers can be isolated from each other. We refer to this system as the helium level valve. A bolometer is used to ascertain the open/closed state of the valve. The pressure in the polarization chamber is monitored with a capacitive pressure transducer, while two carbon resistance thermometers measure the cell temperature. The NMR chamber contains a 285MHz resonator with Q=250. The sensitivity of the NMR spectrometer is 10^{14} fully polarized protons. A millimeter waveguide ends in the NMR chamber making it a Q=1 ESR cavity. The microwave power input into the cavity is adjustable from 1μW to 100μW over the 183GHz to 189GHz frequency range, allowing *b* to *c* conversion rates (and therefore recombination rates) from 10^{15}atoms/s to 10^{17}atoms/s. The *a* to *d* resonance can also be driven, and is used for calibration purposes.

The experiments have been carried out over a temperature range from 0.2K to 0.6K, for both a pure ^{4}He substrate and a ^{3}He-^{4}He mixture substrate. Since the results are similar, we choose to discuss the 0.3K experiments with a ^{4}He substrate. The experiments start with the helium level valve closed. Atomic hydrogen gas

is admitted into the polarization chamber and reaches close to 100% nuclear polarization at about 10^{16}atoms/cc in 30 minutes. The helium level valve is then opened, allowing the doubly polarized atomic hydrogen gas to flow into the NMR chamber. The *b* to *c* transition is driven by the 187GHz ESR, with the *b+c* recombination taking place in the NMR chamber. Pressure and temperature measurements allow monitoring of the recombination process. Pulsed proton NMR (285MHz) measures the polarization of the molecular hydrogen sample during and following the recombination process.

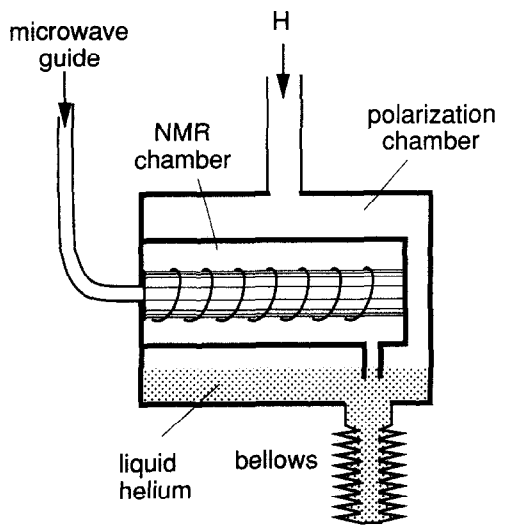

FIGURE 2
Schematic diagram of the experimental cell.

Figure 3 shows a 40minute time sequence of the NMR integrated amplitude. Molecular production rates are 10^{17}molecules/s for this fill. The solid line is a fit to the data by the standard form for the recovery of a saturated spin system to its equilibrium value, using a single relaxation time constant:

$$A = A_0\{1 - \exp[(t_0-t)/T_1]\} \quad (1)$$

For the 0.3K cell temperature, and the 6.7T magnetic field, the thermal polarization is about 3.5% of the full polarization of the protons. The initial nuclear polarization of the sample is 0.0±0.3%. This corresponds to a lower limit of 4K for the initial nuclear spin temperature. The first data point of figure 3 is taken 0.5s after the recombination event which lasts about 1s. Given that the molecules form in a fully polarized state, the low polarization at t=0.5s places an upper limit T_1<0.1s on the initial nuclear spin relaxation time. The fast nuclear spin relaxation and the high spin temperature indicate that the highly vibrational and rotational excited molecules do not dissolve in or adsorb on the liquid helium.

The nuclear spin relaxation constant from the fit in figure 3 is: T_1 = 11±1min, in agreement with measurements by other workers (6), when scaled to the ortho concentration and the temperature of the present experiment. The NMR line-width is 150kHz, consistent with the intramolecular dipolar broadening for the fcc phase of solid hydrogen. From the ortho to para conversion rate of the solid we find the initial ortho concentration to be X_0 = 99+1/-2%, therefore confirming the recombination model. An absolute calibration of the signal amplitude shows that the recombination is completely contained in the NMR chamber. The nuclear spin relaxation rate, the ortho to para conversion rate, and the line-width indicate that the solid hydrogen formed by the stimulated recombination of the polarized atomic gas is a standard compact fcc solid.

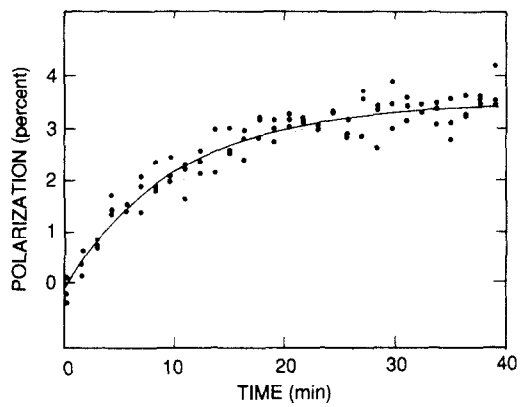

FIGURE 3
NMR integrated amplitude versus time. Sample size in the NMR chamber is $0.7*10^{17}$molecules. Solid line is fit to data using eq 1. T_1 = 11±1min.

In conclusion we have shown that the highly excited molecular gas formed in the recombination process of atomic hydrogen looses nuclear polarization in less than one second. The final product of the recombination is compact fcc 100% ortho hydrogen.

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