STABLE ATOMIC HYDROGEN: POLARIZED ATOMIC BEAM SOURCE

T.O. Niinikoski, S. Penttilä*, J.-M. Rieubland and A. Rijllart
CERN, Geneva, Switzerland

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*) Present address: University of Turku, 20500 Turku, Finland.
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ABSTRACT

We have carried out experiments with stable atomic hydrogen with a view to possible applications in polarized targets or polarized atomic beam sources. Recent results from the stabilization apparatus are described. The first stable atomic hydrogen beam source based on the microwave extraction method (which is being tested) is presented. The effect of the stabilized hydrogen gas density on the properties of the source is discussed.

1. INTRODUCTION

It has been pointed out on several occasions [1-3] that spin-polarized atomic hydrogen gas (H+1) has suitable properties for potential applications in fundamental physics research. In this rather dense gas we have electron and nuclear polarizations, the temperature of the gas is low, and the properties of the hydrogen atoms can be accurately calculated to the relativistic limit. Also, we have a rather good understanding of the physical parameters of the H+1 system. The feasibilities and uses of stable atomic hydrogen have already been discussed [1-4] but until now no experimental applications have been attempted. At CERN, a study of stable atomic hydrogen has been carried out with a view to possible applications in polarized targets or polarized atomic beam sources.

The electron-spin polarized atomic hydrogen gas has been predicted to stay in a gaseous state down to absolute zero temperature, because of the light atomic mass and because the triplet pair-interaction potential has only a very weak attractive part with no possibility of bound states. At low temperatures a scattering of two hydrogen atoms in the triplet state is predominantly s-wave and the scattering length is \(a_s = +0.72 \, \text{Å}\). In the spin-polarized atomic hydrogen gas, \(a_s\) represents an effective hard-core diameter of an atom, and the atoms should then form a close-to-ideal gas.

The principal goal in the recent development of stabilization techniques for atomic hydrogen has been to reach the degenerate gas regime where, according to statistics, the gas might show dramatic behaviour. In this quantum regime the mean thermal de Broglie wavelength \(\lambda = h/\sqrt{(2\pi \, \text{mk}_B T)}\), which is a measure of the average spatial extent of the wave packet of a hydrogen atom, begins to be the size of the mean spacing \(d = n^{-\lambda/3}\) between particles; here \(n\) is the density of the atoms. Lhuillier and Laloe [5] have predicted that already in a non-degenerate and nuclear spin-polarized gas, when \(\lambda \leq a_s\), there exist exchange processes which have effects on the nuclear-spin transport properties. When the mean free path of atoms is large enough, it is possible to observe collective nuclear spin oscillations—spins waves. At a temperature of 0.3 K the thermal wavelength is 32 Å, and with a density of \(10^{17} \text{ cm}^{-3}\) the mean spacing is 220 Å. We are now in the classical regime.

The H+1 gas is stabilized against very reactive recombination processes by making the electron-spin Boltzmann factor, \(\exp (\mu B/k_B T)\), very large, which guarantees that the two upper hyperfine states |c\> and |d\> are not thermally populated. For this we need low temperatures (0.2-0.6 K) and a high magnetic

*) Present address: University of Turku, 20500 Turku, Finland.
field (≥ 5 T). The walls of a stabilization cell and the magnetic field gradients will confine hydrogen atoms. Condensation on the walls is reduced by means of superfluid ⁴He and ³He/⁴He linings, which have absorption energies much below those of ordinary solid materials such as metals or solidified gases. On superfluid ⁴He the absorption energy is about 1 K, and on a ³He-coated wall it is 0.3 K.

The highest densities of H⁺ at present exceed 10¹⁸ cm⁻³, observed in bubbles of 100 μm diameter after compression [6-8].

2. STABILIZATION APPARATUS

The CERN stabilization apparatus shown in Fig. 1 consists of a powerful horizontal dilution refrigerator [9], a warm-bore superconducting solenoid of 5 T, and installations for electron paramagnetic resonance (EPR) experiments at 140 GHz and continuous-wave nuclear magnetic resonance (CW–NMR) at 1 GHz. At room temperature, dissociated hydrogen atoms are fed into the cryostat through the heated PTFE transport tube, which is 6 mm in diameter and 90 cm in length. In the transport tube the atoms thermalize through wall collisions. The gradient of the solenoid field spin selects and confines the atoms. The cold part of the transport tube is lined with helium film. The apparatus has a loading rate of over 10¹⁶ stabilized atoms per second into the sample cell. At a magnetic field of 5 T and a temperature of below 0.5 K we have measured densities up to 10¹⁷ cm⁻³. The sample cell area arrangement inside the mixing chamber is shown schematically in more detail in Fig. 2. The copper parts have been fixed to the accommodator shown in Ref. [10]. However, Fig. 2 does not show the ³He–NMR coil in the ³He–⁴He mixture for tuning the magnetic field.

Total pressure of the H⁺ gas was determined by means of a 10 mm diameter, capacitive Mylar membrane pressure gauge, which was part of a 53 MHz tunnel-diode oscillator circuit. In the measurements the third harmonic was counted. Calibration, which was done with the helium vapour, gave a sensitivity of Δf/Δp ≈ 3 x 10⁶ Hz/Torr.

The 1 GHz resonator was made from a copper wire, 50 μm in diameter and 6 mm long. The wire was mounted to the end of a coaxial line and was located within a cylindrical sample volume, 15 mm long and 5 mm in diameter as shown in Fig. 2. The CW–NMR measurements were performed by sweeping the frequency. The circuit uses a hybrid bridge and a double balanced mixer for coherent homodyne detection [11].

The volume of the EPR cavity is about 7 mm³; compared with the previous experiments [10], we expected a significant reduction in the observed EPR linewidth. The H⁺ atoms are confined between two windows in the ends of the cylindrical-shaped cavity, which communicates through a narrow channel with the load and stabilization volume. The 0.8 mm diameter and 10 mm length of this channel were chosen in order to attenuate the 140 GHz microwave field strongly in this structure well below the cut-off of propagation. The magnetic field was swept slowly across the resonance line, and the density decay, caused by recombination of flipped atoms, was monitored by means of the pressure sensor. A bolometer, weakly coupled to the microwave field, allowed the intensity of the microwave field to be monitored. From an I–V curve the responsivity of the bolometer was calculated to be ΔR/ΔP ≈ 5 x 10¹⁰ Ω/W at 0.3 K.

3. RESULTS

Resonance measurements were done in the magnetic field around 5 T, limited by the band of our carcinotron microwave source. A higher field would reduce recombination and relaxation rates. In our configuration the volume/surface ratio is constant, because the volume of the H⁺ gas is limited by the field gradients and helium-coated walls. Such a quantity of helium gas was condensed onto the surfaces that the moving helium film did not cause any significant pressure oscillations in the loading tube. Such oscillations were seen to affect the loading speed.
Figure 3 shows the time evolutions of the pressure and the CW-NMR absorption signal area at a temperature of 0.28 K and a magnetic field of 4.9 T during and after a fast loading. The upper part shows the pressure on logarithmic scale and demonstrates the density decay according to the rate equation

\[ \dot{n}/n^2 = K + 2G, \]  

(1)

where \( K \) is the surface recombination rate constant and \( G \) is the rate constant for nuclear relaxation from the state \( |b\rangle \) to the state \( |a\rangle \). In the case of Fig. 3, \( G \) includes also relaxations induced by the RF field.

The ground hyperfine state \( |a\rangle \) is the mixed spin state, which has a small electron spin-up admixture. The state \( |b\rangle \) is the pure spin state. Thus collisions involving an \( |a\rangle \)-state atom can result in recombination to molecular hydrogen at a rate which is higher than the nuclear relaxation rate. After closing a valve in the filling tube, recombination on the surfaces reduces the density rapidly as long as there are enough \( |a\rangle \)-state atoms. The surface recombination rate constant of \( K = 3 \times 10^{-18} \, \text{cm}^3/\text{s} \) is obtained from the fast part of the decay. After depletion of the \( |a\rangle \) atoms, the nuclear relaxation rate constant \( G \) starts to control the decay. The fitting to the linear part of the curve gives the relaxation rate constant of \( G \approx 8 \times 10^{-20} \, \text{cm}^3/\text{s} \); the corresponding nuclear polarization is \( P \approx [1 - (G/K)] \approx -97\% \) during that part of the time evolution.

In Fig. 4 we see a typical averaged NMR absorption signal at a temperature of 0.3 K, and at the field of 5.065 T which corresponds to the resonance frequency of 929 MHz. The sweep width of the frequency was 320 kHz and the density of the sample about \( 10^{16} \, \text{cm}^{-3} \). To average one signal we needed about 15 s. The homogeneity of the field in the NMR volume was determined and tuned by condensing \(^3\)He gas to the volume and measuring the frequency of the \(^3\)He NMR. The \(^3\)He NMR linewidth gives an effective homogeneity of \( 1 \times 10^{-4} \). The \( H \rightarrow \) NMR signal width from Fig. 4 gives \( \Delta B/B \approx 4 \times 10^{-4} \), which is about four times larger than that of the \(^3\)He signal. This is caused by the nuclear-spin transport phenomena in the NMR volume [12]. This is also supported by the clearly observable fine structure on the top of the absorption signal, persisting over long averaging periods. Details of the fine structure can be better seen by averaging the sweeps in only one direction instead of averaging them in both directions.

The spin waves split the nuclear magnetic resonance into peaks spaced at

\[ \omega - \omega_0 = \mu PD_z k^2 \]  

(2)

from the Larmor frequency \( \omega_0 \); here \( k \) is the wave number of the mode in the spin wave resonator, \( \mu \) is the ‘identical spin rotation’ parameter [5], and \( D_z \) the diffusion coefficient for the transverse magnetization [5]:

\[ D_z = D_0 /[1 + (\mu P)^2]. \]  

(3)

The longitudinal (ordinary) spin diffusion coefficient \( D_0 \) is given by [5]

\[ D_0 = (3/8)(k_BT/nm)(1/\Omega_n^{1,1}), \]  

(4)

where \( n = n_a + n_b \), \( m \) is the mass of the \( H \) atom, and \( \Omega_n^{1,1} \) the relevant collision integral [5]. Our observed line shapes depend on the polarization qualitatively according to Eq. (2), although the spin wave modes \( k \) cannot be clearly resolved. A more complete account will be published elsewhere.

In the absence of long-range spin-spin interactions, the longitudinal nuclear polarization of the two level system, defined by \( P = (n_a - n_b)/(n_a + n_b) \), is proportional to the area of the CW-NMR
absorption signal which depends on the microscopic complex susceptibility \( \chi = \chi' + i\chi'' \) of the sample. The absorption signal area in a linear system should be \( S = \int \chi''(\omega) \, d\omega \propto P \cdot n \). The requirement that \( |P| \) should stabilize at 0.97, when the density decay becomes controlled by the nuclear relaxation, is clearly violated by our data shown by the dots representing \( s/n \), the signal surface scaled by the atomic density (given by the pressure signal).

The effects of the transverse spin diffusion on the signal surface \( S \) are very small in the absence of losses of transverse magnetization, although the height and width of the spin wave peaks depend on \( D_z \) in first order. The losses of transverse magnetization result from dephasing in the gradient of the external field or on the surfaces, from magnetic relaxation on surfaces, and from exchange of atoms through the filling hole. In the simplest picture we may assume that the transverse magnetization in the rotating frame is inversely proportional to the diffusion coefficient: \( \chi''(\omega) \propto D_z^{-1} \). This gives immediately

\[
S \propto P \cdot n \cdot D_z^{-1} \propto P \cdot n \cdot D_0^{-1} \propto P \cdot n^2
\]

using Eqs. (3) and (4). Figure 3 shows also the polarization evolution \( P \propto S/n^2 \), which gives a much better fit to the required asymptotic saturation of \( P \) towards the value \(-0.97\).

A more complete discussion on the NMR signal surface area will be published elsewhere [13].

The EPR detection of the H\textsubscript{1} atoms was based on the deduction of a part of the sample through recombination during partial saturation. One advantage of the small cavity, which was about 10 mm away from the axis of the solenoid field, was that the resonances \( |a\rangle \rightarrow |d\rangle \) and \( |b\rangle \rightarrow |c\rangle \) can be approached from the high- or the low-field side. The pressure sensor was used to measure the line width, which was about 20 G, and the calculated gradient of the field within the EPR cavity at 5 T was about 100 G/cm. The resonance absorption was not strong enough for detection by the bolometer. Only weak EPR-induced recombination heat was observed. With the estimated microwave power level of 50 nW, \( 5 \times 10^{15} \) spin flips per second could be monitored when the field was swept at 2 G/s over the resonance.

4. **STABLE ATOMIC HYDROGEN BEAM SOURCE**

It has been shown that the most promising use of stable atomic hydrogen can be as a polarized atomic beam source [10]. Stabilized atoms are extracted by flipping the magnetic dipole moments of polarized hydrogen atoms by means of the EPR microwave field. Because of the topology of the solenoid field, a large fraction of the atoms will flow out of the magnet without recombining during collisions. At a high density the motion of the flipped atoms is diffusion—or drift—driven by the gradients of the solenoid field. The collisions which limit the motion cause relaxation, recombination, and nuclear depolarization due to spin exchange.

4.1 **CERN source**

The source in construction and tests consists of a loading tube and storage volume, surrounded by the dilution refrigerator and the storage magnet. Additional solenoid, or hexapole magnet optics can be installed to increase the beam density in the beam detectors, which include a compression tube and ion gauge, and a quadrupole mass-spectrometer with crossed-beam ionizer. The source will be pulsed by microwave power modulation.

Figure 5 shows the polarized atomic hydrogen source which is being tested at CERN. The basic apparatus is the same as in the stabilization experiments. The accommodator and stabilization assembly has been replaced by a large copper tube traversing the mixing chamber. The open end of the tube forms an extraction orifice. The short PTFE tube ends at the still, where warm (~ 30 K) hydrogen atoms will thermalize on the cold surfaces coated by superfluid helium. A loading rate of \( 10^{18} \) per second has been achieved so far. Before the storage volume there is a low-density buffer of unpolarized atomic hydrogen.
gas. Because of the open configuration, special care has been given to the helium film. Burning and condensing of the film are carried out in the still and the heat exchange region where most of the cooling power is available.

The position of the extraction orifice can be changed with respect to the field of the 5 T solenoid. The length of the solenoid is 180 mm. The inner diameter of the storage cell is 25 mm. The effective storage volume is about 130 cm\(^3\), and if the density in the middle of the 5 T field is \(n_0 = 10^{16} \text{ cm}^{-3}\) there are \(3 \times 10^{17}\) trapped H\(\perp\) atoms.

4.2 Microwave extraction

The electron spin of the H\(\perp\) atom is reversed adiabatically when an atom crosses the resonance surface. The time which the flipped atom then needs to get out of the magnet (and then also the flux of atoms) depends strongly on the density and the gradients in the resonance region. In low densities, \(n < 10^{15} \text{ cm}^{-3}\), when the mean free path is approximately equal to the diameter of the storage volume, the beam quality is determined by the source temperature and focusing solenoid optics. At higher density the motion of the flipped atoms is limited by interatomic collisions. In a field gradient the motion is directional and is characterized by a drift constant. In constant field the flipped atoms move randomly obeying the law of thermal diffusion. For the purpose of the following simplified discussion, we shall ignore interatomic collisions at a density below \(10^{15} \text{ cm}^{-3}\); only wall collisions are then limiting the number of atoms emerging into the beam tube. Track-tracing simulations have been performed [14] to follow the free motion of the atoms in this low-density limit.

In order to find an optimum working density and the resonance region for a required pulse length \(\tau_p\) and duty cycle, we have estimated the drift speed \(v_0\) of a flipped atom along the symmetry axis of our magnet. The \(v_0\) is a function of the field gradient \((\nabla |\mathbf{B}|)_z\), of the temperature, and of the diffusion coefficient \(D_0\) [5] which depends inversely on the density. The origin is in the middle of the solenoid magnet, where the field was 5 T and the temperature of the H\(\perp\) gas was 0.3 K. In Fig. 6 is shown the flow time \(t_f\) of flipped atoms as a function of the \(z\) coordinate for various densities \(n_0\) in the centre of the solenoid (where \(z = 0\)). Also, the field dependence of the mean free path of H\(\perp\) atoms, \(\ell = 1/(n\sqrt{T})\), is shown by the dashed curve when \(n_0\) is \(10^{16} \text{ cm}^{-3}\). The resonance (spin-flip) points on the axis are \(z = 20\) mm and \(z = 40\) mm. When \(z = 20\) mm the field and the corresponding axial gradient are \(\mathbf{B} = 4.949\) T and \((\nabla |\mathbf{B}|)_z = -460 \text{ G/cm}\), respectively. When \(z = 40\) mm the values are \(\mathbf{B} = 4.823\) T and \((\nabla |\mathbf{B}|)_z = -920 \text{ G/cm}\). Figure 6 clearly shows how sensitive \(t_f\) is to the density and the field gradients. When \(n_0 = 10^{16} \text{ cm}^{-3}\) an atom which leaves from \(z = 20\) mm spends 80 ms reaching point \(z = 60\) mm, whilst an atom leaving \(z = 40\) mm needs only 17 ms to get to the same point.

In the pulsed operation, if the length of the microwave pulse is \(\tau_p = 2\) ms and the duty cycle is 10%, the working density should be about \(3 \times 10^{15} \text{ cm}^{-3}\) so that \(t_f\) would be smaller than \(10 \times \tau_p\) when the leaving area is \(z = 20\) mm.

The model can also be used to estimate the probability for an atom in the hyperfine state \(|d\rangle\) or \(|c\rangle\) to leave the magnet in its original state when it is moving in the H\(\perp\) gas.

A contribution of the three-body recombination was calculated with the rate constant estimated by Bell et al. [15]. There is a 97% probability that the atom will survive when the density is \(n_0 = 10^{16} \text{ cm}^{-3}\) and it has left from the resonance point \(z = 20\) mm.

The two-body collision of two atoms in different electron-spin states on a superfluid helium-coated surface is very destructive. Owing to the topology of the solenoid field, the two-body recombination rate is insignificant. When \(z = 20\) mm and we are 12 mm from the axis, the radial gradient is \((\nabla |\mathbf{B}|)_r = +57 \text{ G/cm}\), which will drive the atoms toward the axis.

The effect of the electron-spin dipole relaxation was estimated by using the result of Kagan et al. [16]. In the above conditions the probability is about 95% that the atom will drift to the lower density area without the electron-spin relaxation.
From an earlier discussion on the electron-spin exchange [10], the exchange cross-section $o^{ex}$ can be used. This gives the same spin-exchange-rate constant ($T_{1}^{-1} = 0.54 \times 10^{-12}$ s⁻¹) as has been calculated by Morrow and Berlinsky [17]. The spin-exchange rate is so large that the nuclear polarization of the beam will be close to zero even at low energies.

In the following we consider a flux to an interaction channel, of diameter 10 mm, to build up a density of $10^{15}$ cm⁻³. When the source temperature is 0.3 K the average velocity after acceleration of the 5 T field is 250 m/s. This gives the required current density of $3 \times 10^{17}$ s⁻¹ cm⁻². In a pulse of 2 ms we then have a total of $6 \times 10^{14}$ atoms, and if the duty cycle is 10% then the average flux is $3 \times 10^{16}$ s⁻¹. If now the efficiency of our source is 10% (including focusing of atoms, recombinations, relaxations) we need the flipping rate of $3 \times 10^{17}$ atoms per second.

To reach this time-scale the resonance surface has to be in a region of strong field gradient. To produce so many adiabatic reversals of electron spins at the resonance surface, the microwave field strength must be high enough [10]. The microwave power level now depends on a field gradient $\nabla |\mathbf{B}|$, an amplitude of an alternating field $B_1$, and the thermal speed of the atoms. Input power is related to the size and the quality factor of the EPR volume.

5. CONCLUSION

The stable atomic hydrogen experiments have supported and also more precisely limited the concepts of applications discussed here, most of which are based on the pulsed neutral beams available with microwave extraction.

With the stabilization apparatus, we have achieved a high loading speed of $10^{16}$ H⁺ per second into the storage cell and, after the modification, up to $10^{18}$ H⁺ per second have been detected. This is required for a large throughput of hydrogen atoms in a source.

The CW-NMR experiments on H⁺ gas have shown that during high-speed loading the $|a\rangle$ state is more populated than it would be in thermal equilibrium. The NMR signals have also shown the existence of the nuclear-spin transport in H⁺ gas. The EPR experiments—at least in a small cavity—have proved that in a field gradient of 60 G/cm without any high microwave field strength, large adiabatic spin-flip rates are possible.

The first working refrigerator suitable for a stable atomic hydrogen source experiment with the open storage cell configuration is ready and is being tested.

The properties of the source depend very much on the working density. When the density is higher than $10^{15}$ cm⁻³, a diffusional motion determines the minimum length and maximum duty cycle of the beam. By placing the resonance region in a strong gradient ($\nabla |\mathbf{B}|) = 600$ G/cm, and working at a density under $10^{16}$ cm⁻³, it seems possible to form pulses of a few milliseconds length with a 10% duty cycle and to obtain beam densities of more than $10^{13}$ cm⁻³ at the focusing point.

The electron spin-exchange cross-section is so large that the nuclear polarization in the beam will be almost zero and a RF transition is needed downstream in the beam in order to convert the electron polarization into nuclear polarization.
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Fig. 1  CERN stabilization apparatus. The main parts are a dissociator, a powerful horizontal dilution refrigerator, and a superconducting solenoid of 5 T with a warm bore.

Fig. 2  Schematic of the sample cell configuration inside the mixing chamber.
Fig. 3  Time evolutions of the total density, probed by the pressure gauge, and of the NMR absorption signal area.

Fig. 4  Typical CW-NMR absorption signal, after averaging 800 sweeps, at a field of 5 T and temperature of 0.3 K. The density of the sample was about $10^{16}$ cm$^{-3}$.
Fig. 5  CERN H⁺ source.

Fig. 6  Mean flow-time of H⁺ atoms along the symmetry axis of the solenoid field in dense H⁺ gas. The two resonance regions $z = 2$ cm and $z = 4$ cm are in the field gradient area. The density in the middle of the magnet is a parameter. Also, the field dependence of the collision mean free path on the field is shown as the dashed line when $n_0 = 10^{16} \text{cm}^{-3}$ at the origin.