

Production and Storage of Polarized H₂, D₂ and HD Molecules

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The investigation of the recombination of nuclear-polarized hydrogen and deuterium atoms into polarized molecules gives new insights into different fields in physics and chemistry. On the one hand, the measurement of the polarization preservation during the recombination process itself allows one the observation of a new parameter during the chemical processes on different surfaces and enables the production of hyper-polarized molecules. On the other hand, the molecular polarization, measured as a function of the imposed external magnetic field, delivers the coupling forces of the nuclear spins and the rotational magnetic moments of the molecules as well as the coupling between the two nuclear spins themselves. This knowledge allows one to optimize storage-cell gas targets for accelerators experiments or to produce and handle polarized fuel for future fusion reactors.

(The corresponding talk was also given at the PSTP2017 conference in Kaist, South Korea [1]. Nevertheless, this proceeding includes recent developments and more technical details in subsection 2.1.)

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1. Introduction

Experiments with polarized internal gas targets have shown that the nuclear polarization of hydrogen and deuterium atoms, produced with polarized atomic beam sources, is at least partially preserved in the recombination to molecules on different surfaces [2, 3, 4]. During the recombination process, the properties of the surface determine the nuclear polarization of the molecules. For example, it was shown that a water surface prevents the recombination process [5] and metal surfaces made from copper or gold restrict the nuclear polarization to about 50% of the incoming polarization of the atoms [3, 6]. In this case, a possible explanation is the Eley-Rideal mechanism: A polarized atom, reaching the wall, will be adsorbed by a covalent binding with the metal atoms. Either due to the electron exchange or due to the undetermined magnetic moments of the free electrons in the metal, the nucleon in the adsorbed atom will depolarize very fast. A second polarized atom from the gas can recombine with the depolarized one without ever been adsorbed on the surface. Then, only the nuclear polarization of the second atom is preserved, which is responsible for the nuclear polarization of the molecule. Of course, the electron spins in the molecule must be anti-parallel aligned. This means that the absence of free electrons will decrease the recombination rate. Therefore, the knowledge of the achieved molecular polarization and the recombination rate of hydrogen and deuterium atoms in different hyperfine substates (HFS) allows one to investigate the recombination process on surface materials.

Besides the polarization loss during the recombination process, the molecules will also lose polarization due to the spin-spin interaction between the nuclear spins I_1 and I_2 itself or due to the spin-rotation interaction between the nuclear spins I_1 and I_2 , coupled to I , with the rotational moment J of the molecule. For example, during the wall collisions, m_J can change and, therefore, transitions between different m_I states occur, *i.e.* nuclear polarization is lost. An external magnetic field B can reduce this coupling to m_J . In Ref. [6] the formula to describe this dependence is given as

$$P_m(\bar{n}, B) = \frac{P_{m_0}}{1 + (B_c/B)^2 (\bar{n}/\ln 2)} \quad (1.1)$$

Here, $P_m(\bar{n}, B)$ is the nuclear polarization of the molecules in a storage cell with an average number of wall collisions \bar{n} in a magnetic field B . The so-called critical field B_c corresponds to the magnetic field seen by one of the two nuclei due to the rotational moment J and the spin I_2 of the other nucleus. It corresponds to the coupling power between J and I , and I_1 and I_2 .

In an ensemble of hydrogen molecules, the occupation numbers of the rotational states depend on the temperature. Below 200 K all hydrogen molecules with parallel nuclear spins (ortho-hydrogen) are in the lowest $J = 1$ state, and at room temperature of about 300 K only 12% are in the first excited rotational state $J = 3$. For ortho-hydrogen (both nuclear spins are coupled to $I = 1$), due to the necessary anti-symmetric wave function of this ensemble of fermions, only odd $J = 1, 3, 5 \dots$ values are allowed. Para-hydrogen ($I = 0$) must have even J . From the known occupation numbers, the critical field can be calculated. For hydrogen, *e.g.*, below 200 K it is $B_c = 5.4$ mT and at 300 K it is increased to 6.5 mT. This means that polarized hydrogen molecules ($m_I = \pm 1$) can never have $J = 0$ and, therefore, the polarization of the molecules can never be maintained in the gas phase for a long time. When hydrogen molecules are cooled down to liquification temperature, J must become $J = 0$ and the ortho-hydrogen molecules will change into unpolarized para-hydrogen.

For deuterium molecules the situation is opposite: In this ensemble of bosons the $I = 2$ ortho-deuterium state, with the perfectly vector-polarized substates $m_I = \pm 2$ can have $J = 0$ at low temperatures around 40 K. In addition, the spin-spin coupling is much weaker. Thus, polarized deuterium molecules might be stored at large magnetic fields for a reasonable time and it should be possible to freeze them out as polarized ice.

To investigate the recombination processes on different surfaces, the polarization losses due to wall collisions, and the polarization lifetime of the molecules, a collaboration of the St. Petersburg Nuclear Physics Institute (PNPI), the Institute for Nuclear Physics of the University of Cologne and the Institute for Nuclear Physics of the Research Center Jülich was established to develop a dedicated experimental setup.

2. The Apparatus

The polarized atomic beam source (ABS), formerly used to feed the polarized internal target of the ANKE experiment at COSY/Jülich [7], is used to produce a beam of hydrogen or deuterium atoms in HFS with selected nuclear and electron spin orientations. The polarized atoms enter a T-shaped storage cell (see Fig. 1), where they can recombine into molecules on dedicated surfaces. The storage cells can be exchanged to enable measurements with different inner surface coatings on the fused-quartz wall. The temperatures of the storage cell can be set between 45 and 120 K due to the contact to the liquid helium cooling of the superconducting magnet and a heater at the center of the cell. The superconducting solenoid allows one to set a magnetic field in the storage cell to values up to 1 T. In addition, this magnetic field helps to focus the electron beam from the electron gun through the storage cell. Due to electron impact at energies of a few 100 eV the polarized atoms and molecules are ionized. By this, molecular ions H_2^+ can only be produced from the molecules, but protons can stem from atoms or from molecules. The cross sections for the ionization processes are well known [8]. Afterwards, the ions are accelerated by an electric potential on the cell and focused into the Lamb-shift polarimeter (LSP). The kinetic energy of the ions is fixed by the potential of up to 5 keV. With non-conducting surface materials, the field gradient is produced by the current in a set of gold-plated tungsten wires stretched along the cell.

The Lamb-shift polarimeter [9] is a perfect tool to measure the nuclear polarization of protons/deuterons or the molecular ions $\text{H}_2^+/\text{D}_2^+$ [10]. First, a Wien filter with its crossed magnetic and electric fields is used as a velocity filter to separate the different ions. In addition, the perpendicular magnetic field induces a Larmor precession of the proton/deuteron spin. By choosing the corresponding field strength the precession can be fixed to 180° so that the nuclear polarization is inverted. This effect does not influence the molecular ions, because due to the larger magnetic moments of the electrons their spins precess much faster and, therefore, they are reoriented adiabatically along the external magnetic field. The nuclear spin is coupled strongly to the electron spin and its orientation follows. Behind the Wien filter, both atomic and molecular ions reach the cesium cell. By charge exchange with the cesium vapor inside a strong magnetic field metastable atoms in dedicated HFS of the $2S_{1/2}$ state are produced. The only difference for both types of ions is the efficiency of their neutralization processes that is smaller by a factor of about 35 for the molecular ions. The spinfilter, the next component, is able to transfer all metastable atoms into the ground state. Only at special resonant conditions of an RF field, a static electric and a magnetic

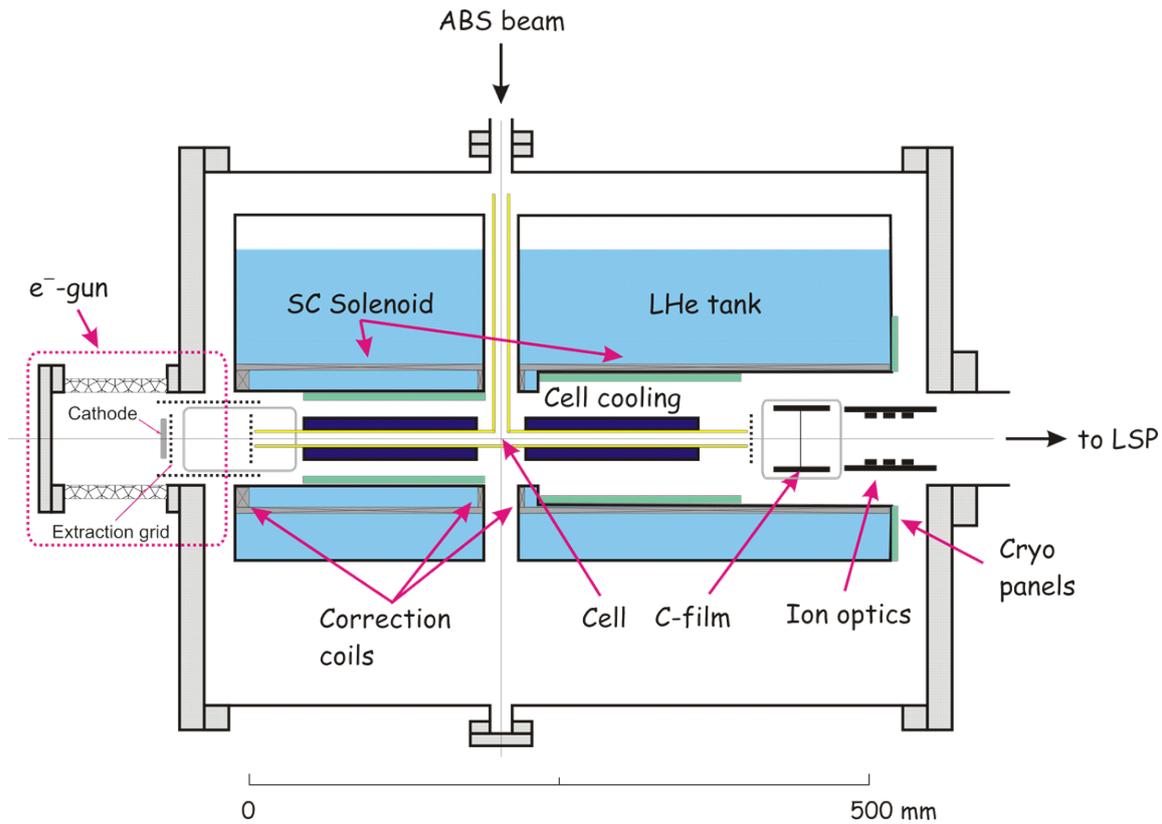


Figure 1: The interaction chamber: polarized atoms from the ABS enter the exchangeable T-shaped storage cell, where they can recombine into molecules. Inside a strong magnetic field of up to 1 T, produced by a superconducting magnet at 4 K, the atoms and molecules are ionized by an electron beam entering from the left. The ionized protons and H_2^+ ions are accelerated by a positive electric potential on the cell of up to 5 kV and focused into the LSP on the right side to measure the nuclear polarization.

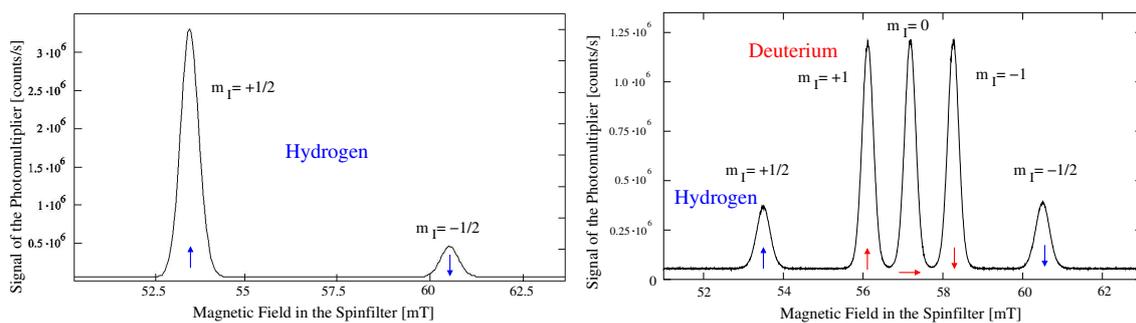


Figure 2: On the left side: a typical Lyman- α spectrum of positively polarized protons. The polarization can be determined by comparing the asymmetry of the signals at 53.5 mT ($m_I = +1/2$) and 60.5 mT ($m_I = -1/2$) and adding some minor correction factors. In this example the nuclear polarization of the protons was $P_z = 0.78 \pm 0.01$. On the right side: a Lyman- α spectrum of unpolarized protons and deuterons. At 55.5 mT metastable deuterium atoms with a nuclear spin projection of $m_I = +1$ can pass the spinfilter, those with $m_I = 0$ at 56.5 mT, and at 57.5 mT those with $m_I = -1$.

field, metastable atoms survive and pass the spinfilter. These metastable atoms are quenched into the ground state and the produced Lyman- α photons are registered with a photo-multiplier as a function of the magnetic field strength (see Fig. 2). The ratios of the peaks contents with small corrections yield the polarization of the beam from the storage cell.

2.1 Technical Details

During these experiments several technical problems appeared. The following list represents some dominant aspects that are important for the understanding of the measurements.

1. The preparation of the surface

Due to the necessary positive potential of the storage cell, the conducting surface must be electrically isolated from the support system. In addition, the storage cell is cooled via this support system made from copper, which has thermal contact to the liquid helium reservoir of the superconducting solenoid. Therefore, the storage cells itself are made from fused quartz tubes with a wall thickness of 2 mm and an outer diameter of 14 mm. This type of glass allows large temperature gradients and potentials up to +5 keV of the inner surface compared to the support system.

An inner metal surface is produced by vaporization of, e.g., a thin gold wire (5 μm) that was coiled around a tungsten wire of 0.1 mm a few hundred times. For this reason, a dedicated vacuum chamber was built with a special support system to keep the expanding tungsten wire in the center of the storage cell during the heating process with currents up to 50 A. This method works for most metals, but especially nickel was a problem for a long time, because liquid metal embrittlement of the tungsten wire due to the liquid nickel destroyed the wire and did not allow the necessary vaporization temperatures. This problem was finally solved by use of a thicker tungsten wire and higher currents for the heating process. Furthermore, the amount of the surface material has to be rather small, because the adhesion of the material on the fused quartz can keep not more than a few 100 nm on the wall. The metallic surface thickness has a resistance about 1 Ω along the cell. This resistance can be used to produce a small electric field along the cell when the surface is set on a potential of about 1 or 2 keV. This additional longitudinal field accelerates the positively charged ions inside the cell into the direction of the Lamb-shift polarimeter and helps to increase the intensity of the outgoing beam. To set the necessary electric potential of the cell for non-conductive surfaces, e.g. the fused quartz itself or Fomblin oil, four thin (5 μm) gold-plated tungsten wires are placed along the cell and fixed on both ends. These positively charged wires create a quadrupole field that focuses the ions radially along the center of the storage cell and helps to increase the intensity of the ion beam. Even for metal surfaces, these wires are often used, because they offer an easier way to contact the inner surface for the necessary potential.

2. Build up of a water layer

When a surface is cooled down in vacuum, it will start to adsorb water from the residual gas and build up a water layer on top. This effect was used by the HERMES collaboration [5] to cover their storage-cell surface with a water layer, because water is up to now the best known surface material to avoid the recombination of the hydrogen atoms and preserves the nuclear polarization of the hydrogen atoms down to temperatures of 80 K. At lower temperatures

the polarization gets lost and is gone at about 40 K due to the increase of the dwelling time, i.e. exactly in our temperature regime. Thus, if polarized molecules shall be produced, a water surface has to be avoided as much as possible. For this reason, the small amount of oxygen was canceled, which had been given to the dissociator of the ABS to increase the degree of dissociation and the intensity of the atomic hydrogen beam. Some of the water, produced in the dissociation plasma, would be still in the ABS beam. The volume around the storage cell is pumped via cryo-panels on the liquid helium tanks of the superconducting magnet to reduce the partial pressure of water. A cooled cryo-trap between the ABS and the recombination chamber should avoid a diffusive water flux from the ABS. In addition, the storage cell is heated to 310 K after implementation into the apparatus, when the surrounding components are cooled down to liquid nitrogen temperature. Nevertheless, depending on the surface and the vacuum conditions, even in the best case it only takes a few days until the cold surface is covered with water. In this case, a current of a few A can be sent through a metal surfaces to heat it to temperatures of about 400 K for a few minutes to get rid of the water. Due to the good heat isolation of the fused quartz 2 to 20 kW are enough for this purpose, so that the surrounding components are not heated too much. The temperature of the surface itself can be measured due to the increasing resistance during the heating process. In a previous experiment Price and Haeberli [12] measured the nuclear polarization of hydrogen atoms as function of the temperature on different surfaces and observed for most of the surfaces the same temperature dependence as for water. From our point of view it is now obvious that they got a water layer on top when they cooled down their cells. In corresponding experiments with a gold surface, in our set-up the nuclear polarization of the molecules did not decrease at temperatures between 100 and 45 K. Therefore, the polarization of the atoms before the recombination did not depend on the temperature in this regime any more.

3. Zero-crossings of the magnetic fields

When the spin of a nucleus is coupled to the spin of an electron, the polarization of both particles is very sensitive to zero-crossings of the magnetic field. The much faster Larmor precession of the electron allows these particles to follow modifications of the magnetic field direction very fast. If these particles pass a region of zero magnetic field, the nucleus is depolarized very quickly. This effect was observed at two different parts of our apparatus:

a.) When the polarized atomic beam enters the reaction chamber, the beam has to pass the superconducting magnet. Along the vertical feeding tube of the storage cell inside the magnet there is a region, where the magnetic field along the inner part of the solenoid is compensated by the outer magnetic field to close the magnetic field lines. The magnetic shielding of the reaction chamber itself is absorbing external fields. To avoid a zero crossing of the atoms a small additional solenoid was placed on top of the chamber to produce a vertical magnetic field. The current to drive this solenoid had to be tuned according to the magnetic field along the cell. To avoid this complicated procedure, we found that the persistent current switch (PCS) of the superconducting solenoid can be used to overcome this zero-crossing. The PCS is placed on top of the solenoid, but still in the liquid helium bath. The magnetic field of its superconducting wire covers the zero-crossing region for magnetic fields up to 1 T. Only at 1 T itself the additional vertical solenoid has to be used.

b.) When the polarized ions are leaving the reaction chamber, they have to pass through a gate valve and an electrical lens before they reach the magnetic field of the Wienfilter. In this region no dedicated magnetic field was defined, because the decreasing longitudinal magnetic field of the superconducting solenoid should change smoothly into the transverse field of the Wienfilter, so that no zero-crossing was expected nor observed for several years. Nevertheless, the exchange of the gate valve was producing a zero-crossing due to magnetic screws inside the new valve. The polarization of the protons/deuterons was not influenced, but the much more sensitive H_2^+ ions, still containing one electron coupled with the nuclear spins, were partially depolarized compared to former measurements. A new gate valve with stainless steel screws solved this problem.

3. The Results

Typical results of polarization measurements of the protons and H_2^+ molecular ions are shown in Fig. 3. As described by formula 1.1, the polarization decreases with decreasing magnetic fields along the storage cell. The left panel shows the polarization of protons, extracted after ionization from a gold-coated cell kept at 44 K and fed by hydrogen atoms in the HFS 1 ($m_I = m_j = +1/2$). The protons can be produced either from molecules or from atoms. Therefore, formula 1.1 must be modified to

$$P_p(B) = a \cdot P_a^0 \cdot f_{\text{HFS}}(B) + b \cdot \frac{P_{m_0}}{1 + (B_c/B)^2 (\bar{n}/\ln 2)} \quad (3.1)$$

Here P_a^0 is the polarization of the hydrogen atoms from the atomic beam source and $f_{\text{HFS}}(B)$ describes the nuclear polarization of atoms in the single hyperfine substates as function of the magnetic field B . $f_{\text{HFS}}(B)$ is equal to one for the pure hyperfine states 1 and 3. The parameters a and b with $a + b = 1$ result from the fit to the measured distribution $P_p(B)$. The degree of recombination c , defined as the fraction of protons in the molecules, is

$$c = \frac{2 \cdot b}{2 \cdot b + a/5}. \quad (3.2)$$

The factor 1/5 takes into account that the electron-impact cross section to ionize a hydrogen atom is by a factor 5 larger than the cross section to produce a single proton from a H_2 molecule.

The (magenta) fit by the function of Eq. (3.1) would allow $c = 1$, *i.e.*, 100% recombination. Due to the lack of data at low magnetic fields, however, a small fraction of free atoms in the cell gas is possible. It would contribute a field-strength independent component in the measured distribution (fit parameter $a \neq 0$).

The average number of wall collisions $\bar{n} = 120 \pm 10$, obtained by the fit of $P_m(\bar{n}, B)$ of Eq. (3.1) to the measured data, perfectly fits to Monte-Carlo simulations under the assumption that the molecules undergo elastic scattering with the walls only. More details are found in Ref. [11].

The right panel of Fig. 3 shows results obtained with a Fomblin-oil coated cell kept at 100 K and fed by hydrogen atoms in the pure HFS 3. Here, the polarization of protons from atoms and molecules was measured separately with the use of the method described in Sec. 2. Those from the molecules yield the polarization of $P_{m_0} = -0.84 \pm 0.02$ during the recombination process, thus, very close to the expected maximum value of -0.87 that can be delivered by the ABS. With the

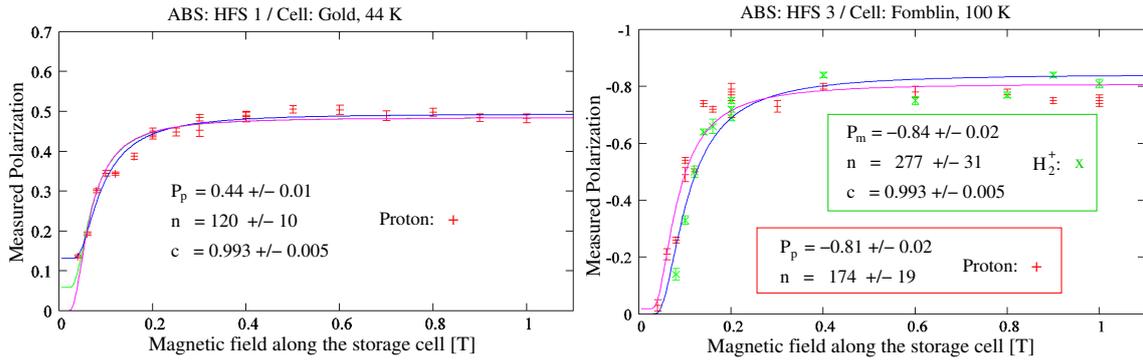


Figure 3: Left side: A typical measurement of the proton polarization after the recombination of hydrogen atoms in the HFS 1 injected into a gold-coated cell kept at 44 K. The fit of Eq. (3.1) yields the average amount of wall collisions as $\bar{n} = 120 \pm 10$. A large recombination of $c = 0.993 \pm 0.005$ can be deduced, because the polarization of the hydrogen atoms in this HFS does not depend on the magnetic field. Right side: A similar measurement on a Fomblin surface. Here, the measured polarization is $P_{m0} = -0.84 \pm 0.02$ for large magnetic fields. Therefore, the nuclear polarization survives the recombination process on this surface.

Wien filter set to the mass-1 separation, one would expect protons from the atomic gas component, at the injection of atoms in the HFS 3 characterized by B -field independence, as well as protons from the ionization of the H_2 molecules. The comparison of the measured dependence on the B -field strength with that of the H_2^+ molecular ions, however, allows a very weak B -field independent component only. Obviously, with the Fomblin-oil surface one achieves an almost complete recombination connected with almost perfect polarization maintenance. This behavior might be explained with the assumption that the atoms are split on the Fomblin surface into protons and electrons. The latter can jump within the bulk of this polymer from one carbon nucleon to another. Therefore, the proton spins cannot interact with the spins of free electrons on the surface and are always aligned with the external magnetic field. Nevertheless, free electrons with an anti-parallel spin are available for the recombination with another atom into the molecule from the bulk. As a result of the fit, the average number of wall collisions for the H_2^+ ions is $\bar{n} = 277 \pm 31$ and, therefore, higher than that for the protons with $\bar{n} = 174 \pm 19$. A plausible explanation for this observation may be found in the different average free path lengths of protons and H_2^+ ions inside the storage cell at an average pressure of 10^{-4} mbar. Protons, produced in the center after a few wall collisions, can leave the storage cell but the larger H_2^+ ions mostly cannot. They interact with the atoms and molecules in the gas and are partially lost. Only those molecular ions that are produced at the end of the cell, after a larger number of wall collisions are extracted into the LSP.

Although the molecular polarization for a Fomblin surface is twice that with the gold surface, the use of gold offers an advantage: Cold surfaces in vacuum will catch water from the residual gas. But a water surface seems to be the best material to avoid the recombination. Depending on the amount of water in the vacuum chamber it takes between hours and up to 2 days until the water surface is dominating. In this case, a measurement on the Fomblin surface must be stopped. Another observed detail was that in this case the recombination rate c depends on the HFS of the atoms. E.g., when hydrogen atoms in the HFS 2 and 3 are sent into the storage cell at the same time, the

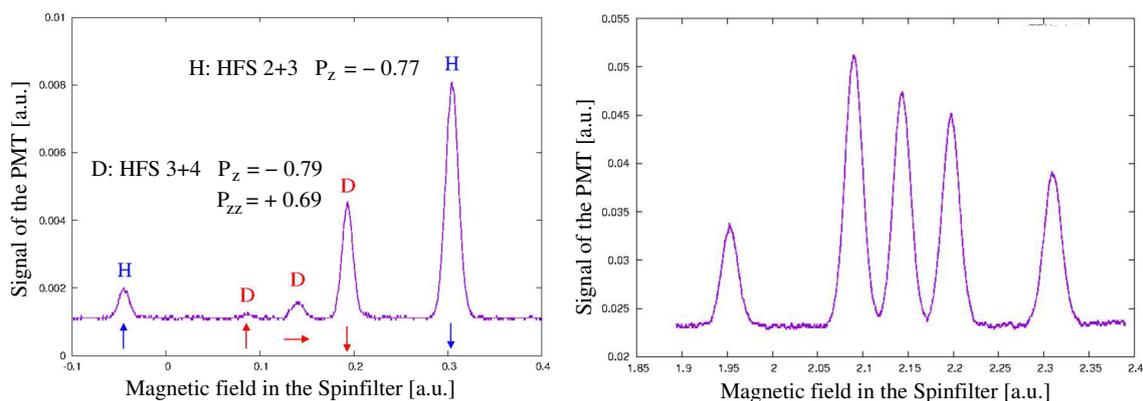


Figure 4: Lyman- α spectra of HD^+ ions measured with the LSP. Polarized hydrogen and deuterium atoms recombined either on a Fomblin surface (left side) or on a gold surface (right side). The nuclear polarization, given to the atoms, can be found in the HD molecules again.

recombination rate was still rather large. But when only atoms in HFS 1 are used, c becomes much smaller. This can be explained with the assumption that the HFS 2 and 3 have the same negative nuclear polarization, but anti-parallel electron spins. Therefore, they can easily recombine into molecules, where anti-parallel electron spins are needed. If now atoms in HFS 1 only are used, it seems that water is not able to offer free electrons with anti-parallel spins.

In contrast to the Fomblin, the gold surface can be heated to 400 K to vaporize the water when a small current of a few A is send through. Thus, the measurements can continue when the storage cell is back at low temperatures after about one hour. In the case of Fomblin oil, this requests replacement by a virginal cell. Therefore, most measurements are made on gold surfaces up to now.

In more recent experiments the ABS was fed with hydrogen and deuterium atoms at the same time. Depending on the setup of the ABS transition units, atomic states of different hyperfine-substate occupation could be injected into storage cells with Fomblin-oil or gold surfaces. In one of the experiments, hydrogen atoms in HFS 2 and HFS 3 together with deuterium atoms in HFS 3 and HFS 4 were injected into a Fomblin-oil coated cell kept at 100 K. All components were characterized by negative nuclear spin orientation. The nuclear polarization in the recombined molecules H_2 , D_2 , and HD was determined with the LSP for mass separation $m = 2$, $m = 3$, and $m = 4$ by the Wien filter. The left panel of Fig. 4 shows the signal of the photo-multiplier as a function of the magnetic field in the spin filter for $m = 3$, the HD molecules. Again, the polarization of both nucleons was very large with $P \sim -0.8$ close to the theoretical maximum. Some small losses come from the not perfect settings of the weak-field transitions unit of the ABS that could not be used for both isotopes perfectly at the same time. In the example on the right side of Fig. 4 a negative polarization was chosen for the protons, but a positive vector-polarization for the deuterons. Here, the gold surface did not allow for large polarization values. But, as can be seen from these examples, with this method it is possible to produce HD molecules in many different HFS combinations.

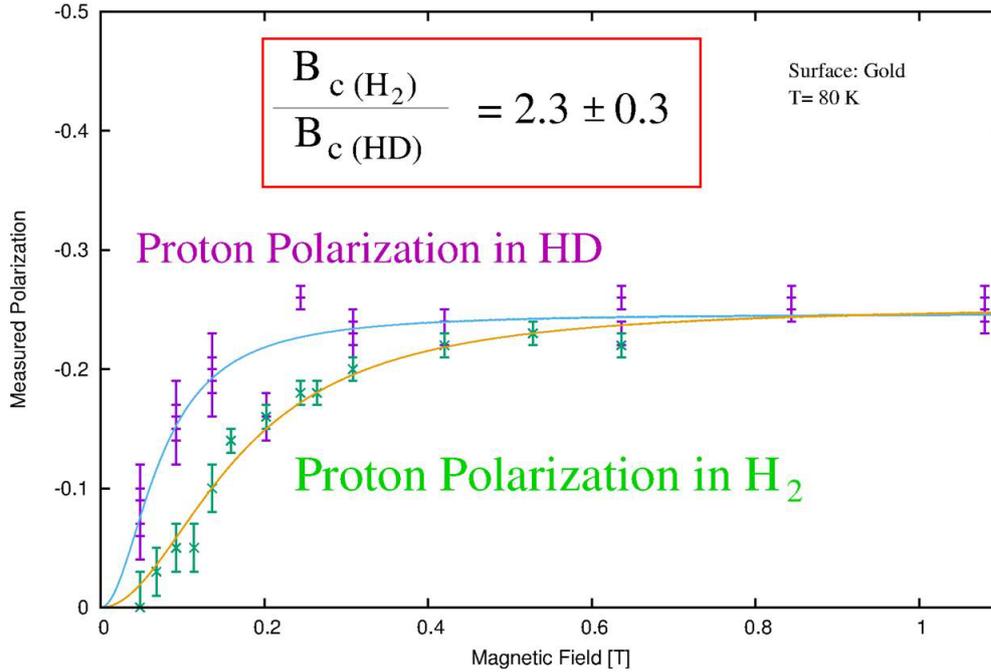


Figure 5: The polarization of the protons in the H_2 (green curve) and in the HD molecules (magenta curve) after recombination of polarized hydrogen and deuterium atoms. From this measurements can be deduced that the coupling between the nuclear spins and the rotational magnetic moment J , *i.e.* the critical field B_c , is not equal.

Besides HD molecules, H_2 and D_2 molecules are created from the injected polarized hydrogen and deuterium atoms. These are symmetric rotators, whereas the HD molecules are asymmetric rotators. The coupling strength of the proton to the rotational moment and the deuteron is different in both cases as it is shown in Fig. 5. The proton polarization in the HD molecule approaches the saturation value at lower external field strengths than that of protons in the H_2 molecule. Therefore, the binding of the proton spin in the HD molecule is weaker than in the H_2 molecule. The strength of the coupling in the fit function of Eq. (1.1) to both measured magnetic-field dependences is determined by the parameter B_c , the critical magnetic field strength. The two fits, performed under the assumption of equal mean numbers of wall collisions \bar{n} , yield $B_c(\text{H}_2)/B_c(\text{HD}) = 2.3 \pm 0.3$.

The ratio can be explained in the classical picture of the rotation of two charged objects of masses m_1 and m_2 , coupled at a fixed distance d . Both objects are characterized by a magnetic moment. The rotation produces a magnetic dipole field B_{rot} , which in the rotational plane decreases proportional to $1/r^3$, where r is the distance to the rotational axis. In the case of H_2 , $m_1 = m_2$ is equal to the proton mass m_p . For both protons the distance to the rotational axis is $r = d/2$ and the coupling of the magnetic moment to B_{rot} is $\propto 1/(d/2)^3 = 8/d^3$. In the HD molecule with $m_1 = m_p$ and $m_2 = m_d = 2m_p$ the distance of the proton to the rotational axis is $2d/3$ and that of the deuteron $d/3$. Here, the coupling of the proton to B_{rot} is $\propto 1/(2d/3)^3 = 27/8d^3$. The ratio $8/(27/8) = 2.4$ for the coupling in H_2 and HD, respectively, agrees with 2.3 ± 0.3 from the fits. In this naive calculation the spin-spin coupling is not included. Again, the coupling between the proton and the deuteron at a distance d is smaller for the HD molecule than between two protons in the H_2 molecule.

4. Conclusion

The possibility to investigate the recombination process of polarized atoms into polarized molecules gives new insights in different fields of physics and chemistry. First of all the behavior of the nuclear spin during a chemical reaction is now observable with this setup. Experiments with several surfaces show a different dependence of the surface materials and allow one to distinguish between various recombination mechanisms.

Also the coupling power between the rotational magnetic moment J and the nuclear spins can be investigated. When the parameter B_c in the fit function of Eq. 1.1 is known, the average number of wall collisions \bar{n} can be determined. Monte-Carlo simulations help to better understand the interaction of the hydrogen molecules with the wall material. *E.g.*, elastic scattering on the surface delivers values of \bar{n} lower than those, which result for molecules kept on the surface and re-emitted with a \cos or even a \cos^2 angular distribution.

This knowledge is helpful for future polarized internal gas targets at accelerators, because:

1. Molecules are slower than atoms at the same temperature. Therefore, they need more time to leave the storage cell, which increases the target density.
2. The experience of the polarized target at HERMES [13] has shown that the storage cell temperature should not be below 100 K for hydrogen atoms to avoid loss of nuclear polarization. The experiments with polarized molecules have shown that lower temperatures are possible without polarization losses. The target density is increased by cooling of the cell to lower temperatures.
3. Molecules can stay on selected surfaces and leave the surface with a \cos or even a \cos^2 angular distribution, which leads to an increased number of wall collisions and an increased target-gas density.

Another application could be to use polarized D_2 , HD or possibly TD molecules as polarized fuel in nuclear fusion reactors. It is well known that the fusion cross sections are spin dependent and can be increased when the fuel particles are polarized. *E.g.*, the $d + t \rightarrow {}^4\text{He} + n$ reaction rate would be increased by a factor of 1.5 if the spins of d and t are aligned [14].

As a next step it is planned to freeze out polarized D_2 and HD molecules on a cooled surface below 10 K inside the strong magnetic field of the superconducting magnet. If the polarization will be preserved long enough, the polarized ice can be used as a polarized target for laser-acceleration experiments to produce polarized deuteron beams. For polarized HD molecules also polarized proton targets are possible. Even laser-induced fusion experiments are in range with polarized solid D_2 or HD ice. Earlier production and use of polarized HD ice targets in photon-scattering experiments are described, *e.g.*, in Ref.[15], [16].

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