PROCEEDINGS OF SCIENCE

Polarized H_2, D_2 and HD molecules and their possible use to feed a polarized H_2^+, D_2^+ or HD^+ ion source for stripping injection into storage rings

R. Engels,[a,b,*] **R. Gebel,**[a,b] **K. Grigoryev**[a,b] **and Y. Valdau**[a,b]

[a] *Institut für Kernphysik, Forschungszentrum Jülich, Wilhelm-Johnen-Str. 1, 52425 Jülich, Germany*

[b] *GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstraße 1, 64291 Darmstadt, Germany*

E-mail: r.w.engels@fz-juelich.de

With a dedicated apparatus it was shown that the nuclear polarization of hydrogen atoms and its isotopes, produced by a polarized atomic beam source (ABS), can be preserved during the recombination into molecules. In this way, polarized H_2 and D_2 molecules in hyperfine substates where both nucleons have the same nuclear spin are generated. In more recent experiments the ABS was used to determine the spin of hydrogen and deuterium atoms passing through in parallel. In this case, the nuclear spins of the protons and the deuterons can be determined separately to get HD molecules in any hyperfine substate, i.e. in any spin combination. One application of this technique can be the design of an intense H_2^+, D_2^+ or even an HD^+ polarized ion source for stripping injection at storage rings like COSY with polarization values above 0.8 and intensities in the 10 μA range.

19th Workshop on Polarized Sources, Targets and Polarimetry (PSTP2022), 26-30 September, 2022 Mainz, Germany

*Speaker

PoS(PSTP2022)008

1. Introduction

Actual polarized ion sources for accelerators are based on two principles: Optical pumping with laser beams or the ionization of the polarized beam of an atomic beam source (ABS) based on the Rabi-Apparatus and the Stern-Gerlach experiment. Highly polarized protons are induced by both methods, but polarized deuteron can be produced with an ABS only. For stripping injection into storage rings normally H_2^+ or D_2^+ are used, but even negative ions, i.e. H^- or D^- are possible, but more complicated to produce. If polarized proton or deuteron beams are foreseen actual storage rings are equipped with polarized negative ion sources only [1, 2], even when in this case more complicated unpolarized H^-/D^- sources must be implemented too. Another disadvantage of this method is the limitation of the beam intensity, because the cross sections of the charge exchange from a polarized atom into a negative ion are smaller than the ionization into protons by electron impact like in an ECR ionizer. Nevertheless, the polarized negative ion sources are the only option for stripping injection, because polarized H_2^+/D_2^+ ions were not available until now.
Polarized atomic beam sources again are used to implement a polarized hydrogen/deuterium target into a storage ring: Either the polarized hydrogen beam of an atomic beam source (ABS) is used as a jet target [3, 4] or this beam will feed a T-shape storage cell target [5–9]. The advantages of the hydrogen jet target are the purity of the atomic beam, a small beam-interaction volume with the accelerator beam and the large polarization of the atoms that can be measured in parallel with a Breit-Rabi [10] or a Lamb-shift polarimeter [11] precisely. But the target density will be limited to 10^{12} cm^{-2} due to the maximum flux of an ABS of about 10^{17} atoms/s. When the atomic beam is used to feed a T-shape storage cell the target density increases up to 10^{14} cm^{-2}, but the interaction volume is increased too. The polarization of the hydrogen atoms in the storage cell can be measured by nuclear reaction until the corresponding analyzing powers or, in combination with a polarized proton beam, the spin-correlation coefficients are known [5, 6]. Another method is to extract a small amount of atoms and measure the polarization with a Breit-Rabi polarimeter [7]. During the development of this storage cells it was obvious that depending on different surface materials the nuclear polarization of the hydrogen inside was decreasing compared to the primary beam leaving the ABS. Of course, any ferromagnetic material must be avoided, because magnetic field gradients on the surface are destroying the nuclear polarization rather fast. In parallel, it was realized that chemical reactions of the radical hydrogen atoms are another source for polarization losses. Especially the recombination into molecules was dominant for many materials, but nevertheless it was shown that the nuclear polarization survived at least partially during the different recombination processes [12–14].

2. The Apparatus

For the development of the polarized internal target at ANKE [15] a dedicated apparatus was built and used to investigate this recombination processes even further [16] and maybe to find a material that is able to preserve the nuclear polarization during the recombination process. In Fig. 1 the schematic setup is shown: The polarized atomic beam from an ABS is sent into a T-shape storage cell where the atoms are stored up to 1 ms and will hit the wall surface depending on the reflection processes more than 100 times before they will leave the cell. To preserve the nuclear

polarization an external magnetic field up to 1 T is needed that is induced by a superconducting magnet around the cell. The liquid helium tanks around the superconducting coil are not just cooling the coil itself, but their outer surface is partially covered with carbon pellets and, thus, they are a powerful cryo-pump. In this way pressures of 10^{-7} mbar are possible even with the full flux of the ABS entering the cell.

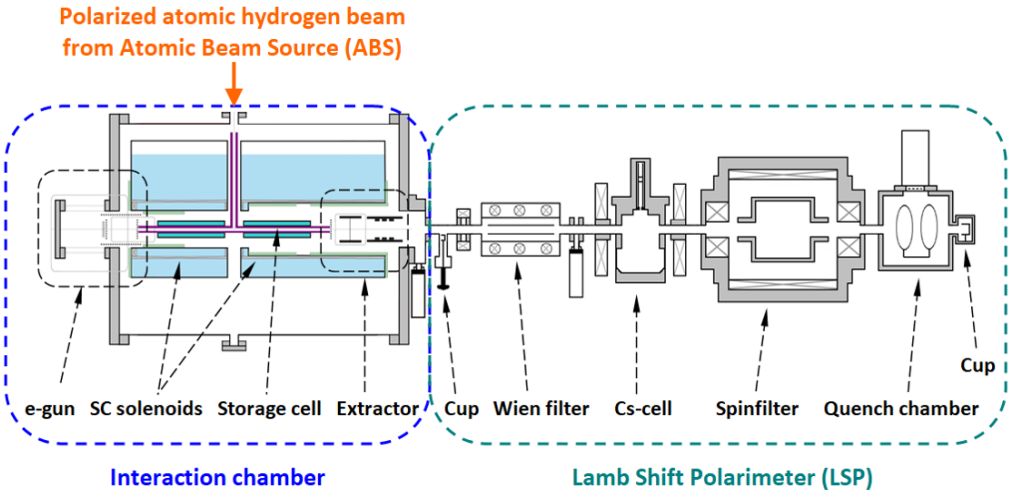

Figure 1: The design of the apparatus to produce polarized molecules and to measure their polarization.

The cells itself are made from fused quartz and are 400 mm long with 14 mm outer and 10.8 mm inner diameter. The temperature of the cell during operation can be regulated between 40 and 120 K. An electron beam of a few μA from an electron gun on the left is send into the cell that can be set to a positive potential up to several keV either via a metallic inner surface or with thin gold-plated tungsten wires along the cell. The longitudinal magnetic field helps to keep the electrons on the inner axis and allows a large electron density inside. Electron impacts will ionize the atoms and molecules and the potential along the cell will accelerate the produced protons/deuterons and the molecular ions at the end of the cell into the following Lamb-shift polarimeter (LSP). Here, the first component is a Wien filter that allows with its crossed electric and magnetic fields to separate the ions due to their different velocities. Afterwards, the polarization of the single nucleons is measured separately, because the LSP can determine the amount of protons/deuterons with different spins. For further details of the LSP see ref. [11]. Even when the Wien filter can not separate between H_2^+ and deuterons, because both particles have mass 2, the LSP can measure the polarization of the nucleons independently like shown in Fig. 2. In this special case the cell was filled with polarized hydrogen and deuterium atoms at the same time and the HD^+ ions are separated with the Wien filter (mass 3) [17]. The inner surface of the cell can be prepared with different methods. For example, experiments are made with gold, copper, aluminum and even ferromagnetic nickel surfaces. As next a carbon coating is foreseen. One of the simplest methods is to build up a surface from fluids like Fomblin oil. Due to the fact that the partial pressure of this polymer is below 10^{-7} mbar even at room temperature it will not vaporize during the operation at temperatures below 100 K.
Another important parameter for the polarization preservation is the magnetic field along the cell.

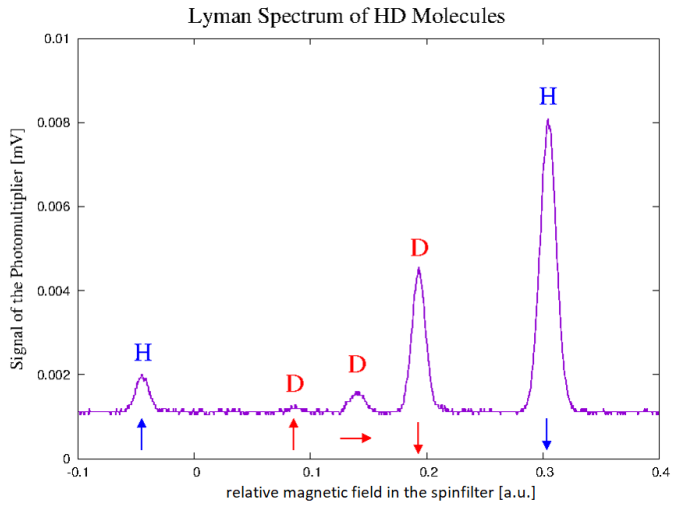

Figure 2: The Lyman spectrum of the LSP for negatively polarized HD molecules after recombination of polarized hydrogen and deuterium atoms on a Fomblin surface: The arrows show the spin direction of the protons (blue) and deuterons (red) at the corresponding magnetic fields in the spinfilter of the LSP. Due to the different peak intensities the polarization of the HD molecules can be calculated to $P_z(\mathrm{H}) = -0.77 \pm 0.01$, $P_z(\mathrm{D}) = -0.79 \pm 0.01$ and $P_{zz}(\mathrm{D}) = +0.69 \pm 0.02$.

Due to the coupling of the nuclear spins I to the magnetic moment of the molecules induced by the rotational momentum J the polarization can be lost during a single wall collisions that might change the direction of J. An external magnetic field B can decrease the influence of J, because the nuclear spins will couple most likely to B. Thus, even a few hundred wall collisions in average will not destroy the nuclear polarization of the molecules. This effect can be investigated by measuring the nuclear polarization as function of the magnetic field B inside the cell like shown in Fig. 3. Measurements like in Fig. 3 allow a deeper insight into the recombination processes on the different

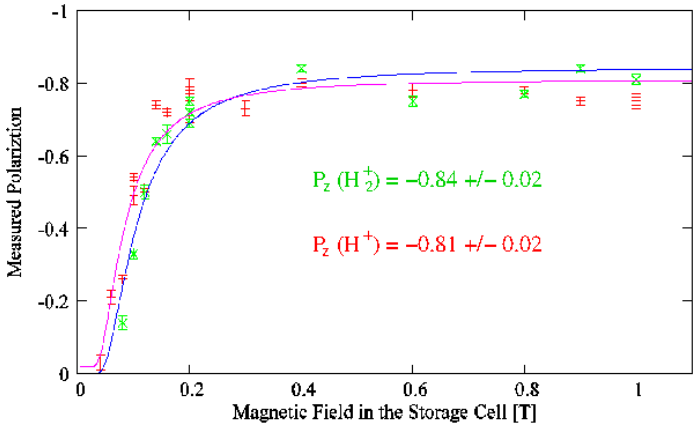

Figure 3: Measurement of the atomic hydrogen (red) and molecular hydrogen polarization (green) as function of the external magnetic field. At magnetic fields higher than 0.3 T the nuclear polarization of $P_z = -0.84 \pm 0.02$ is preserved in the molecules.

surfaces itself. For example, the nuclear polarization of the hydrogen molecules after recombination on a gold or copper surface allow a maximum polarization of $P_z = 0.45$ only, i.e. just 50% of the atomic polarization determined by the ABS. This can be explained by a complete polarization loss of an atom when it will be bound on the surface before it will recombine with another still polarized atom from the gas phase. Futher magnetic impurities at the surface can even decrease the maximum possible polarization.
On a Fomblin surface the polarization is fully preserved which hints to a different recombination process: The proton and the electron of a hydrogen atom are separated on the surface and the proton is captured in potential traps before it recombines with another atom from the gas phase. Otherwise, the hyperfine coupling to the much stronger electron spin would destroy the nuclear polarization via hyperfine interactions. The polarization dependence of the protons and the H_2^+ is very similar. Therefore, most protons must be produced from the molecules and not from hydrogen atoms, because the polarization of the hydrogen atoms in the hyperfine substate 3 ($m_J = -1/2$, $m_I = -1/2$) does not depend on an external magnetic field. Thus, the recombination rate on Fomblin is very close to 1. Even the interaction of the molecules with the surface materials can be deduced, because the coupling between the rotational magnetic moment and the nuclear spins depends on the external magnetic field, but the amount of wall collisions depend on the interaction mechanism between them. More details can be found in ref. [16].

3. A polarized molecular ion source

Beside the investigations of the recombination processes on different materials a modified version of this apparatus can be used to produce an intense and highly polarized H_2^+, D_2^+ or even HD^+ ion beam. The actual setup allows to reach a constant beam with an energy of a few keV, intensities up to 20 μA and polarization values above $P_z > 0.8$. But with an optimized beam extraction and higher voltages of the storage-cell potential even intensities close to a mA might be possible. For this purpose it is very helpful that the recombination rate on a Fomblin surface is very large and that the polarized molecules are stored inside the cell for some time to reach a partial pressure up to 10^{-4} mbar. The cooling of the gas will increase this density even further. That the electron density inside the cell to ionize the molecules is at maximum can be induced with help of the longitudinal magnetic field and the shape of a potential trap that is formed by the positive potential of the cell. Another detail is the larger cross section of the ionization of H_2 molecules compared to atoms. Therefore, the ionization efficiency can be higher than 10^{-3}. Nevertheless, the density of the molecules in the ionization area should not be too large, because the actual measurements have shown that most molecular ions reaching the LSP are produced at the end of the cells, because the average free path length of the molecular ions at this pressures is shorter than the length of the cell itself. For the simplification of such a source it should be mentioned that a superconducting magnetic holding field would be not necessary, because even at 0.3/0.5 T the polarization of the H_2^+/D_2^+ ions reach the saturation like shown in Fig. 3. Another advantage of this kind of ion source would be that the molecular ions still include one electron and its much stronger magnetic moment. In this way, the nuclear spins will be strongly coupled to the electron spin and the electron spin is able to follow changes of the magnetic field direction adiabatically until the gradients are not to strong depending on the velocity of the beam. Thus, the beam transportation is simpler compared

to H^- beams, where the effect of the electrons is canceled and a Wien filter is used to adjust the proton spin direction at the source. Last but not least it must be taken into account that even the stripping cross sections of molecular ions are larger compared to H^- ions and that two protons are produced from one molecular ion instead of one from H^-. Like shown in ref. [17] polarized HD^+ ions can be produced with any spin combination of the proton and the deuteron. This will allow to switch between polarized proton and deuteron beams in the storage ring without major changes at the source. An option that might be useful for coming experiments. In addition, this source can be even used as unpolarized ion source when the cell is just filled with H_2 gas with help of a needle valve.

The only disadvantage identified so far is the attraction of water from the residual gas by the very polar Fomblin surface. In our setup it takes 3 days until the Fomblin surface was covered with water that is the best material to avoid the recombination of the molecules [14]. Of course, the cryogenic pumping helps to suppress the partial pressure of the water and even further pumping will help to avoid this problem and increase the operating lifetime of such a source. Another option might be a regeneration of the surface by spraying some new liquid Fomblin into the cell when needed.

References

[1] A. Zelenski *et al.*; AIP Conf. Proc. **2373**, 070003 (2021).

[2] O. Felden *et al.*; Proc. of the PSP2002 Conf. in Nashville, Indiana, USA, 200 (2002).

[3] H. Rohdjess *et al.*; AIP Conference Proceedings **421**, 99 (1998).

[4] T. Wise *et al.*; Nucl. Instr. Meth. A **556**, 1 (2006).

[5] F. Rathmann *et al.*; AIP Conf. Proc. **421**, 89 (1998).

[6] M. Mikirtychiants *et al.*; J. of Phys. Conf. Series **295**, 012148 (2010).

[7] A. Airapetian *et al.*; Nucl. Instr. Meth. A **540**, 68 (2005).

[8] M.V. Dyug *et al.*; Nucl. Instr. Meth. A **495**, 8 (2002).

[9] P. Lenisa *et al.*; Proc. of the PSTP2019 Conf. in Ferrara, Italy, PoS(PSTP2019) **379**, 25 (2020).

[10] B. Braun *et al.*; AIP Conf. Proc. **421**, 156 (1998).

[11] R. Engels *et al.*; Rev. Sci. Instr. **74**, 4607 (2003).

[12] J. F. J. van den Brand *et al.*, Phys. Rev. Lett. **78**, 1235 (1997).

[13] T. Wise *et al.*; Phys. Rev. Lett. **87**, 042701 (2001).

[14] P. Lenisa *et al.*; Eur. Phys. J. D **29**, 21 (2004).

[15] C. Wilkin; Eur. Phys. J. A **53**, 114 (2017).

[16] R. Engels *et al.*; Phys. Rev. Lett. **115**, 113007 (2015).

[17] R. Engels *et al.*; Phys. Rev. Lett. **124**, 113003 (2020).