











Tests of amorphous carbon-coated storage cells for a polarized gas target at LHCb and further results*

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Abstract: As the LHC beams cannot be polarized, introducing a dense polarized gas target at the LHCb experiment at CERN, to be operated concurrently with beam-beam collisions, will facilitate fixed-target interactions to explore a new energy regime of spin physics measurements. Unfortunately, typical surface coatings used to avoid polarization losses, such as water, Teflon, or aluminum, are prohibited due to restrictions imposed by vacuum and beam policies. The former atomic beam source for the polarized target at ANKE@COSY (Forschungszentrum Jülich), an accompanying Lamb-shift polarimeter, and a storage cell chamber inside a superconducting magnet provide a perfect test stand to investigate the properties of a storage cell coated with amorphous carbon. A significant recombination rate, ranging from 93% to 100%, and preservation of polarization during recombination surpassing 74% were observed. We successfully produced H₂ molecules with a nuclear polarization of $P \sim 0.59$. In addition, we could produce polarized H₃⁺ ions for the first time and observed the shift of the axis of rotation within HD molecules.

Keywords: polarized targets, polarization preservation, storage cell targets, LHC, LHCb

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I. Introduction

After three decades of operation, the Cooler Synchrotron (COSY) at Forschungszentrum Jülich was decommissioned in 2023 [1, 2]. COSY was capable of generating a polarized proton or deuteron beam with a momentum of 3.5 GeV/c. Over a circumference of 184 m, 24 dipole magnets held the pulsed beam, originating from the Jülich Light Ion Cyclotron (JULIC), on its intended trajectory [3]. Sophisticated electron cooling, along with stochastic cooling, was employed to control the beam emittance [4]. An essential part of the accelerator was the ANKE (Apparatus for Studies of Nucleon and Kaon Ejectiles) spectrometer, which allowed for the analysis of ejectiles resulting from collisions between the accelerator beam and a polarized target supplied by an atomic beam source (ABS), as outlined by Mikirtychyants [5] and Bar-

sov [6]. Polarized proton-proton and proton-neutron collisions were of particular interest [7]. Polarization and other properties of the ABS beam were directly assessed using a Lamb-shift polarimeter (LSP) [8] positioned below the ABS [9]. When employed as a jet target, the ABS beam offered a target thickness close to 10¹² cm⁻². Alternatively, a storage cell could be installed within the target chamber to increase the target thickness up to 10¹⁴ cm⁻², thus enhancing the luminosity and facilitating a greater number of double-polarized collisions. The specific properties of these storage cells dictate the characteristics of the provided target. In addition to its length, inner diameter, and temperature that determine the target density, the inner wall coating decides the nuclear polarization of the particles inside.

Currently, ABS and LSP, in addition to an interaction chamber developed through a collaboration between the

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Petersburg Nuclear Physics Institute, the University of Cologne, and Forschungszentrum Jülich [10], operate in a dedicated setup for the investigation of storage cells, as illustrated in Fig. 1. Recent measurements of an amorphous carbon-coated storage cell have shown potential for its use as a polarized target for the LHCspin project within the Large Hadron Collider beauty (LHCb) experiment at CERN [11, 12]. Furthermore, the setup enables the production of H_3^+ ions and facilitates investigations of the rotational magnetic moments of H_2 , D_2 , and HD molecules.

II. EXPERIMENTAL SETUP FOR THE INVESTIGATION OF STORAGE CELL COATINGS

To analyze different storage cell coatings, a polarized atomic beam is introduced into a storage cell, contained in the interaction chamber, via the ABS. The polarization loss and molecular recombination rate of the exiting beam are subsequently measured using an LSP.

Initially, molecular hydrogen or deuterium gas, or a mixture of both, is directed into the ABS dissociator, as shown in Fig. 1, where a resonating radio frequency efficiently dissociates the injected molecules. The resulting atomic gas jet then passes through a sextupole Stern-Gerlach setup, which separates the atoms based on their electron spin. Next, the atomic beam traverses a medium-field transition unit (MFT), where transitions between different hyperfine substates occur. Following this, another set of Stern-Gerlach sextupole magnets is employed. Positioned between these sextupole magnets is a beam chopper, installed to mitigate later interference from Lyman- α photons generated during the dissociation process. Subsequently, the beam progresses to the strong- and weak-field transition units (SFT and WFT, respectively) [13].

Based on the configuration of the Stern-Gerlach mag-

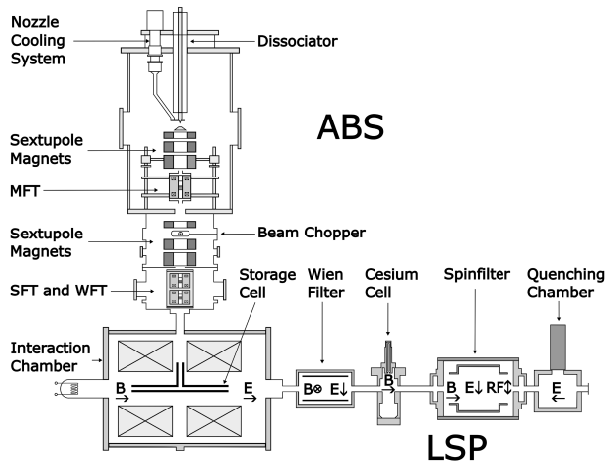


Fig. 1. Schematic of the experimental setup, consisting of an ABS, interaction chamber with a storage cell, and LSP.

nets and transition units, an atomic beam polarized in one or two specific hyperfine substates then enters the interaction chamber. Here, it is guided directly into a fused quartz, a T-shaped storage cell with an inner diameter of 11 mm and length of 400 mm. Atoms colliding with the inner storage cell wall either undergo elastic collisions with the surface or adsorb to it before desorbing according to a \cos or \cos^2 distribution [14]. Depending on the surface material and the beam's polarization, recombination back to H_2 , D_2 , or HD is possible. An electron gun consistently emits a beam of 150 eV electrons into the storage cell to ionize the atoms and molecules, potentially also leading to the dissociation of molecules. Depending on the introduced gas, a mixture of H^+ , D^+ , H_2^+ , H_3^+ , D_2^+ , and HD^+ ions, or just some of these constituents, is produced. Thereafter, an electric potential of approximately 1.5 kV along the storage cell accelerates the ions into the LSP. Superconducting solenoids surrounding the storage cell generate a powerful magnetic field of up to 1 T. The niobium-titanium superconductor wires require cryogenic temperatures below 5.4 K, which are achieved using liquid helium as a coolant. The cell temperature is persistently maintained at 100 K. Pressures within the interaction chamber are of the order of 10^{-8} mbar.

As the ion beam enters the LSP [15], it first encounters a Wien filter, deployed to selectively filter out specific types of ions by exploiting the varying velocities of particles with differing masses. Following this, a cesium cell generates metastable, neutral atoms [16], which then pass through a spinfilter to isolate a single hyperfine substate [17, 18]. Subsequently, a powerful electric field is employed to quench atoms to their ground state (Stark effect). The resulting emission of Lyman- α photons is measured using a photomultiplier (PMT). When the magnetic field of the spin filter is ramped up while hydrogen atoms pass through it, the PMT signal reveals a spectrum with two distinct resonance peaks: one corresponding to hydrogen atoms containing protons with nuclear spin number $S = +1/2$ (at 53.5 mT) and another corresponding to protons with $S = -1/2$ (at 60.5 mT), as illustrated in Fig. 4. By comparing the intensities of these peaks, the polarization can be calculated. In the case of deuterium atoms, three resonances are observed, attributed to deuterium nuclei with $S = +1$ (at 56.5 mT), $S = 0$ (at 57.5 mT), and $S = -1$ (at 58.5 mT).

III. MEASUREMENTS WITH DIFFERENT STORAGE CELL COATINGS

A. Method

The atom density distribution along the storage cell shows a linear decrease with increasing distance from the center, where the beam enters the cell [7]. To enhance the target thickness, various parameters can be adjusted, in-

cluding reducing the cell diameter, elongating the beam tube, and cooling the cell.

The superconducting solenoids encircling the storage cell generate a powerful magnetic field, ensuring that the electron and nuclear spin of hydrogen or deuterium atoms independently couple to the external magnetic field. Despite continuous collisions with the cell's surface coating, the polarization of these atoms persists as long as they remain within a magnetic field of at least a few mT. However, if the coating facilitates the recombination of atoms into their elementary molecules (H_2 , D_2 , or HD), these molecules may lose their nuclear polarization, even in significantly stronger magnetic fields. This is attributed to wall collisions inducing random changes in the molecules' rotational angular momentum projection, which in turn can lead to a transition of the nuclear spin state. Notably, the recombination process itself can result in polarization loss.

According to Wise *et al.* [19], the molecular polarization P_m of H_2 after n wall collisions in an external magnetic field B can be described by

$$P_m(B, n) = P_{m0} e^{-n \left(\frac{B_{c,m}}{B} \right)^2}, \quad (1)$$

where P_{m0} represents the molecular polarization immediately after recombination, and $B_{c,m}$ denotes the critical magnetic field of the molecule. The latter is essentially the strength of the coupling between the molecular angular momentum and total nuclear spin in magnetic field units. For hydrogen molecules with a rotational angular momentum number $J = 1$, the critical magnetic field is $B_{c,m} = 5.4$ mT [19, 20]. It should be noted that H_2 is an ensemble of fermions, and as such, the ortho triplet state of H_2 must have an odd J , whereas the para state must have an even J . At a temperature of 100 K, only low-energy rotational states with $J = 1$ and $J = 0$ are occupied. Here, the para states must have antiparallel proton spins, and only the ortho states can have parallel nuclear spins, i.e., nuclear polarization.

The distribution of the number of wall collisions is characterized by the probability density function $W(n) = \alpha e^{-\alpha n}$, with α being a parameter determined by the surface material, storage cell geometry [21], and wall temperature, which affects the interaction of the molecules with the surface. The mean value of such an exponential distribution is $\tilde{n} = \frac{\ln(2)}{\alpha}$, which can be utilized to calculate the weighted average:

$$\bar{P}_m(B) = \frac{P_{m0}}{1 + \frac{\tilde{n}}{\ln(2)} \left(\frac{B_{c,m}}{B} \right)^2}. \quad (2)$$

If the surrounding magnetic field is strong enough, the ionization process by electron impact preserves the

nuclear polarization. The electron beam produces protons by ionizing hydrogen atoms and can also interact with H_2 molecules, generating both H_2^+ ions and protons. Consequently, when the Wien filter is employed to filter out protons, the polarization of the corresponding ion beam $P_p(B)$ encompasses both the polarization of the fraction a of protons P_a originating from the ionization of their respective atoms and the polarization of the fraction b of those protons arising from the molecules:

$$P_p(B) = aP_a + b\bar{P}_m = aP_a + \frac{bP_{m0}}{1 + \frac{\tilde{n}}{\ln(2)} \left(\frac{B_{c,m}}{B} \right)^2}. \quad (3)$$

Using Eq. (3), an example of the proton polarization for different fractions a is depicted in Fig. 2, showing its dependence on the external magnetic field B applied in the storage cell. The initial molecular polarization was set to $P_{m0} = 0.5$ and the proton polarization to $P_a = 0.9$, with an average number of wall collisions of $\tilde{n} = 200$.

Because two atoms must recombine to build one molecule, the recombination rate c is defined as

$$c = \frac{2 \cdot N_{\text{Molecules}}}{2 \cdot N_{\text{Molecules}} + k \cdot N_{\text{Atoms}}} = \frac{2b}{2b + ka}, \quad (4)$$

where the number of protons produced from molecules b is proportional to the number of molecules $N_{\text{Molecules}}$ in the storage cell. The number of protons produced from hydrogen atoms a must be corrected by a factor $k = \frac{\sigma_{H_2 \rightarrow p}}{\sigma_{H \rightarrow p}} \approx 0.2$ to implement the different cross-sections of the ionization processes.

B. Shift of the rotational magnetic moment in HD molecules compared to H_2 and D_2

The internuclear distance r between protons or deuterons in the H_2 , D_2 , and HD molecule is, to a good approximation, equal. However, due to the mass asymmetry in

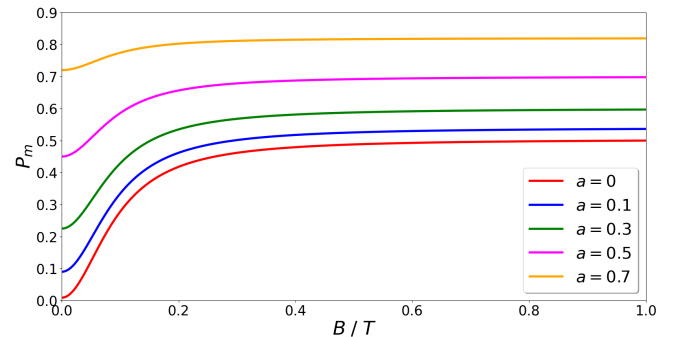


Fig. 2. (color online) Proton polarization for different fractions a as a function of the applied magnetic field B , based on Eq. (3). An initial molecular polarization of $P_{m0} = 0.5$, proton polarization of $P_a = 0.9$, and $\tilde{n} = 200$ wall collisions are assumed.

HD, the center of mass, and thus the rotational magnetic moment μ_J , is shifted towards the deuteron. As a result, the distance d of the proton to the rotational axis increases from $d = r/2$ in the case of H_2 to $d = 2r/3$ in HD. Conversely, the deuteron in HD is located at a distance $d = r/3$ from the rotational axis, in contrast to its symmetric placement in D_2 . When evaluating the coupling strength between the rotational magnetic moment μ_J of a molecule and the corresponding nuclear spins [16, 17], the spatial separation between μ_J , which lies on the rotational axis, and the nuclei is a key factor. For all three diatomic molecules, the same relation for the critical magnetic field B_c holds: $B_c \sim r^{-3}$. Accordingly, a comparison of the proton and deuteron polarization in these molecules as a function of the external magnetic field reveals a modified critical field $B_{c,p}$ for HD, assuming a constant average number of wall collisions n for each of the three molecules. Fig. 3 presents the results for protons (upper panel) and deuterons (lower panel). The expected ratio of the critical fields for protons in H_2 and HD

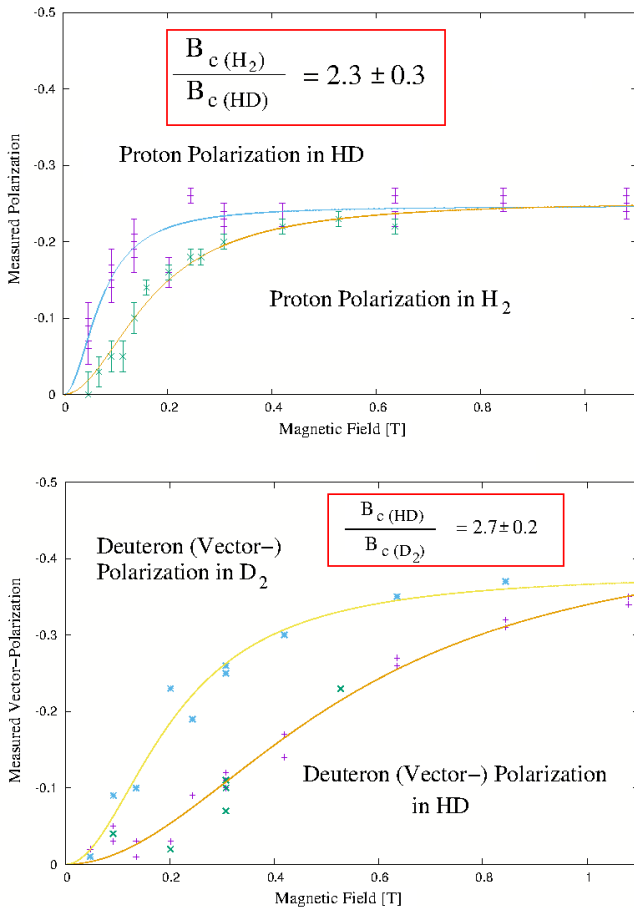


Fig. 3. (color online) Measurements of molecular polarization as a function of the external magnetic field make it evident that the critical field B_{c,H_2} for protons is higher in H_2 molecules than in HD. In contrast, for deuterons, the critical field is higher in HD than in D_2 . This behavior is due to the shift of the rotational axis towards the deuteron in HD molecules.

is

$$\frac{B_{c,H_2}}{B_{c,HD}} = \frac{(2/r)^3}{(3/2r)^3} = 64/27 \approx 2.37. \quad (5)$$

This result agrees well with the measured value of 2.3 ± 0.3 .

For deuterons in HD and D_2 , the expected ratio is

$$\frac{B_{c,HD}}{B_{c,D_2}} = \frac{(3/r)^3}{(2/r)^3} = (3/2)^3 = 3.375, \quad (6)$$

which exceeds the observed value of 2.7 ± 0.2 . This discrepancy may be explained by considering the fact that the assumption of an equal μ_J across all molecules is an oversimplification, as μ_J depends on the rotational level J of the molecules. At $T = 100$ K, nuclear-polarized ortho- H_2 molecules predominantly occupy the $J = 1$ state, while polarized ortho- D_2 molecules are found in either $J = 0$ or $J = 2$. HD molecules, being neither systems of bosons nor systems of fermions, can simultaneously populate states with $J = 0, 1$, and 2.

C. Results for different coatings

When a sufficiently strong magnetic field is applied, the loss of polarization resulting from wall collisions is effectively mitigated, leaving the recombination mechanism, which determines the initial molecular polarization P_{m0} , as the primary process leading to a loss of polarization. Recombination of hydrogen or deuterium atoms can occur through various surface-catalyzed mechanisms, with the Langmuir-Hinshelwood mechanism, hot-atom mechanism, and radiation-induced desorption being the most dominant [22].

In the past, a wide range of commonly used coatings have been investigated in terms of the recombination rate and nuclear polarization preservation that they enable. These studies employed a magnetic field of adequate strength to prevent polarization loss from collisions, with measurements conducted within a storage cell maintained at approximately 100 K. While no perceivable difference in the recombination behavior between hydrogen and deuterium was observed, it is noteworthy that this may vary for different temperature ranges.

Among these coatings, water (or water-ice) has emerged as an optimal candidate for impeding recombination while retaining up to 90% of the polarization of the incoming atoms [14]. However, its unsuitability for vacuum environments poses a significant drawback. Similarly, aluminum and titanium coatings effectively prevent recombination, as both materials are chemically inert. They allow for a polarization preservation of 75%–80% but are characterized by a high secondary electron yield. Additionally, aluminum is known as a reflector for Lyman- α photons with a wavelength of 121 nm. Teflon shares

comparable characteristics in that respect but tends to accumulate charges locally because of its non-conductivity. Gold and copper coatings are recognized as effective catalysts for recombination due to the Eley-Rideal mechanism. However, the preservation of polarization during recombination is fundamentally limited to 50%, as one atom is fully absorbed onto the coating, losing its polarization before recombining with a free atom that retains its polarization [21, 23, 24].

Fomblin oil has shown remarkable properties. As one of the most inactive substances, it exhibits a polarization preservation and recombination rate of close to 100%. However, a significant drawback is its tendency to accumulate water over time, leading to water coverage on the surface of a Fomblin-coated storage cell after approximately three days of operation. In our apparatus with a Fomblin coating at 100 K, a recombination rate of $c = 0.993 \pm 0.003$ and average number of wall collisions of $n = 148 \pm 22$ were observed. Moreover, polarization measurements showed that we achieved atomic and molecular polarizations of $P_{z,p} = -0.81 \pm 0.02$ and $P_{z,H_2^+} = -0.84 \pm 0.02$, re-

spectively [21].

D. Production of polarized H_3^+ ions

In storage cells enabling a high recombination rate, a measurable amount of H_3^+ ions can be built via the reaction $H_2^+ + H_2 \rightarrow H_3^+ + H$. As described in Section 3.1, these stable ions are accelerated into the Wien filter, where they are separated from other ion species in the beam (see Fig. 5). Subsequently, their polarization can be determined from the PMT signal as a function of the applied magnetic field strength in the spin filter. Figure 4 presents such a measurement for H_3^+ ions formed in a gold-coated storage cell. The measured polarization of $P_{z,H_3^+} = -0.41 \pm 0.02$ indicates that a substantial fraction of the initial polarization was preserved during the formation process, as the polarization of the molecules was limited to $P_{z,H_2^+} \sim -0.45$.

Due to the low yield of H_3^+ ions from the source and limited efficiency of the charge-exchange process in the cesium cell, the resulting signals were weak and accompanied by a considerable background. Nevertheless, it was successfully demonstrated that the Lamb-shift polarimeter is capable of measuring the proton polarization within H_3^+ ions.

IV. POLARIZED TARGETS FOR THE LHCb EXPERIMENT

The proposed integration of a polarized gas target for the LHCb experiment is intended to facilitate polarized fixed-target collisions at center-of-mass energies of up to 115 GeV, thereby enabling a wide range of advanced spin physics experiments. The LHCspin project aims to replace the current unpolarized gaseous fixed-target System for Measuring Overlap with Gas (SMOG2) [25] with an ABS alongside a storage cell, providing a dense, polarized atomic or molecular gas target for interaction with the LHC beam. Consequently, it is essential that the coating of the storage cell maintains polarization within the

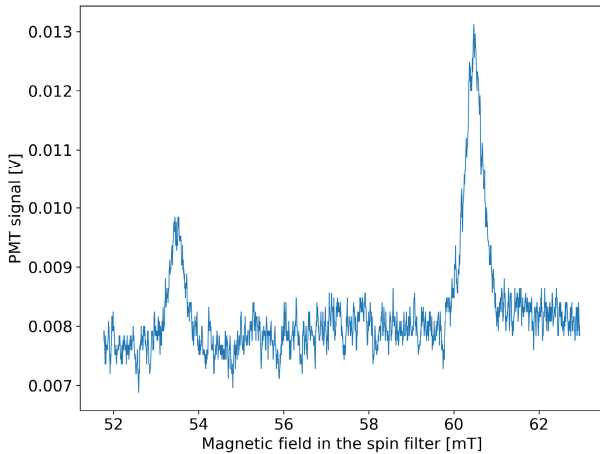


Fig. 4. (color online) The PMT signal as a function of the applied spin filter magnetic field strength for a quenched, polarized H_3^+ beam through the Wien filter reveals a polarization of $P_z = -0.41 \pm 0.02$.

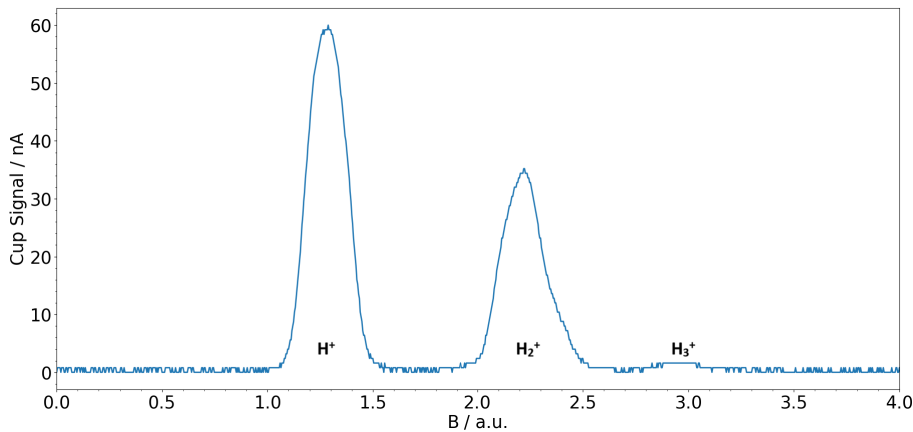


Fig. 5. (color online) Mass spectrum of an ion beam containing H^+ , H_2^+ , and H_3^+ as a function of the magnetic field of the Wien filter.

stored atomic gas while also meeting other crucial requirements. These include a low rate of outgassing to ensure compatibility with vacuum conditions, as well as a minimal secondary electron yield and absence of ferromagnetic properties. In pursuit of this objective, we conducted comprehensive measurements to explore the feasibility of employing an amorphous carbon-coated storage cell to deliver a fixed polarized hydrogen target [26].

Initially, the Wien filter was employed to analyze the various constituents of the hydrogen beam traversing the LSP, along with their respective intensities. This mass spectrometry provides valuable insights into the recombination behavior. In Fig. 5, a measured spectrum is depicted, revealing distinct peaks corresponding to H^+ , H_2^+ , and H_3^+ . The prominent H_2^+ -peak indicates a considerable recombination rate, given that a portion of the recombined H_2 molecules will undergo dissociation, contributing to the number of protons within the adjacent H^+ -peak.

Considering the proton's Larmor precession, which is defined by the angular frequency $\omega_L = -\gamma B$ with gyromagnetic ratio γ and magnetic field strength B , is crucial for minimizing polarization losses induced by the Wien filter along the z -axis. Over a duration τ , this precession will cover an angle of $\beta_L = \omega_L \tau$. When entering the Wien filter, the proton undergoes a transition from a longitudinal to transversal external magnetic field. The proton's weak magnetic moment is not able to follow these changes adiabatically and adjusts its orientation only gradually while traversing the Wien filter. Depending on the time of flight through the device, the proton will exit with different orientations of its magnetic moment, leading to subsequent polarization loss as the magnetic moment realigns itself along the longitudinal direction. Only when the following condition is met – where the magnetic moment undergoes a 180° rotation – will the polarization be fully preserved on the z -axis:

$$\beta_L = \omega_L \tau = -\gamma B \tau = \pi. \quad (7)$$

As shown in Fig. 6, the Wien filter current I_{WF} necessary to induce this rotation was found by assessing the polarization $P_z(I_{WF})$ for various magnetic field strengths. In this case, the resulting data points can be modeled by a cosine function:

$$P_z(I_{WF}) = 0.44 \cos(0.46 \cdot I_{WF} + 3.57). \quad (8)$$

Using this information, we achieved a maximum polarization of -0.64 ± 0.02 when metastable hydrogen atoms were generated in the LSP. Subsequently, we conducted measurements to determine the molecular polarization, attained by filtering for H_2^+ ions, which were then dissociated and converted into metastable hydrogen

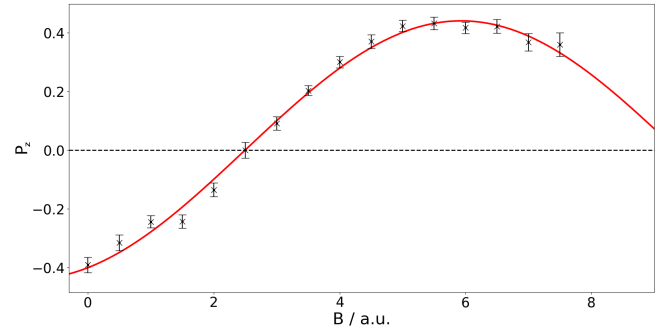


Fig. 6. (color online) The Wien filter curve illustrates the change in polarization P_z measured by the LSP along the longitudinal direction when the external radial magnetic fields generated by the electric current I_{WF} change, which is due to the Larmor precession of the magnetic moment of the proton.

atoms inside the cesium cell. This resulted in a molecular polarization of -0.59 ± 0.02 . Given the known initial polarization of incoming atoms at approximately 0.8, and accounting for the fact that approximately 3.5% of particles entering the storage cell are unpolarized hydrogen molecules, it can be inferred that over 74% of the polarization is retained during the recombination process.

In the next step, we measured the proton polarization for various magnetic fields within the storage cell, analogous to the measurement depicted in Fig. 3. Once again, Eq. (3) was utilized to fit the data and determine the proton fractions a and b , along with the number of collisions \tilde{n} . Using the previously obtained molecular polarization value of -0.59 ± 0.02 as the initial polarization [27, 28], we derived a recombination rate of $c = (96.5 \pm 3.5)\%$ from the approximated value of $a = 0.16 \pm 0.18$. The number of wall collisions was estimated to be $\tilde{n} = 400 \pm 300$, albeit with considerable uncertainty due to the challenges of conducting measurements with weak magnetic fields, resulting in a lack of data within this range.

Remarkably, throughout several weeks of operation, there was no significant accumulation of water on the amorphous carbon surface at 100 K, as such a layer would have hindered recombination.

V. CONCLUSION

Storage cells offer a target thickness increase of two orders of magnitude compared to conventional jet targets. This substantial enhancement translates to significantly improved statistics for a variety of experiments. Depending on the coating and strength of the external magnetic field, storage cells can retain the polarization of the inserted atomic gas, even as the atoms recombine into elementary molecules. Hence, they have proven their utility in the ANKE spectrometer at COSY and are set for future deployment in the LHCb experiment at CERN.

Our measurements have confirmed the efficacy of an

amorphous carbon-coated storage cell providing a polarized H_2 target. Nearly complete recombination of atomic hydrogen gas into H_2 was achieved, while preserving a significant portion of the initial polarization, exceeding 74%. Furthermore, the carbon-coated surface displayed great compatibility with vacuum environments, as demonstrated by the absence of water accumulation over

an extended period spanning several weeks.

It was found that the Lamb shift polarimeter is capable of measuring the nuclear polarization of H_3^+ ions. This made it possible to demonstrate for the first time that these ions, which are formed during the interaction of polarized H_2^+ ions with H_2 molecules, preserve their nuclear polarization throughout the formation process.

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