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Measurement of the nuclear polarization of hydrogen and deuterium molecules using a Lamb-shift polarimeter

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Lamb-shift polarimeters are used to measure the nuclear polarization of protons and deuterons at energies of a few keV. In combination with an ionizer, the polarization of hydrogen and deuterium atoms was determined after taking into account the loss of polarization during the ionization process. The present work shows that the nuclear polarization of hydrogen or deuterium molecules can be measured as well, by ionizing the molecules and injecting the H_2^+ (or D_2^+) ions into the Lamb-shift polarimeter. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4897479]

I. INTRODUCTION

Lamb-shift polarimeters (LSP) are routinely used to measure the nuclear polarization of hydrogen or deuterium beams of polarized atomic beam sources or of ion beams from polarized proton or deuteron sources at energies up to several keV.[1–5] Up to now, a LSP was not employed to measure the nuclear polarization of hydrogen or deuterium molecules. With the development of polarized gas-storage cells, used as internal targets in particle acceleration and storage rings to increase the luminosity, such measurements encountered increased interest. Several experiments, performed at accelerators with the use of nuclear reactions, have shown that a fraction of the nuclear polarization of the hydrogen or deuterium atoms in the gas of a storage cell is maintained in the molecules upon recombination of polarized atoms on the cell walls.[6–8] It had been suggested to measure the nuclear polarization of recombined molecules by extracting the ionized molecules from the cell and accelerating them into a LSP.[9] This method, however, was not applied up to now. Systematic studies of the recombination process were postponed in favor of optimizing the properties of the cell surface to minimize recombination and loss of nuclear polarization of the particles on the walls of the cell.

To study the recombination processes, a setup was developed in close collaboration between the Laboratory of Cryogenic and Superconductive Technique of the Petersburg Nuclear Physics Institute (PNPI) in Gatchina, Russia, the Institut für Kernphysik of Forschungszentrum Jülich, and the Institut für Kernphysik of Universität zu Köln in the framework of a project supported by the International Science and Technology Center (ISTC).[10] The setup, shown in Fig. 1, is operated in combination with the polarized atomic beam source[11] and the LSP,[3,4] developed at Jülich for double-polarized internal target experiments.[12]

The installation allows one to study the recombination of polarized atoms in exchangeable storage cells with different inner surfaces inside a superconducting coil producing a longitudinal magnetic field up to 1 T. Parts of the cold surface of the liquid helium tank are covered by activated charcoal panels for cryogenic pumping to reach pressures $\leq 10^{-8}$ mbar. The temperature of the cooled storage cell can be varied between 40 and 120 K by controlled heating. Atoms and recombined molecules in the cell gas are ionized by a $\sim 1\mu$A electron beam of 50 to 150 eV, produced by an electron gun and focused along the cell axis. The protons (or deuterons) produced from atoms and from dissociative ionization and the H_2^+ (or D_2^+) ions are extracted and accelerated by an electric potential of up to +8 keV, applied to the cell, and focused into the LSP. The LSP components are discussed in Sec. II.

The original intention was to strip the residual electrons from the H_2^+ (or D_2^+) ions in a thin carbon film (see Fig. 1) placed in the magnetic field of the solenoid to maintain the polarization of the two emerging protons (or deuterons). Behind the foil the energy of the protons (or deuterons), stemming from the atoms in the cell gas, would be twice the energy of the protons (or deuterons) from dissociative ionization of the extracted molecular ions, which have to share the energy. The protons (or deuterons) from the molecular ions could be prevented from reaching the LSP by setting the carbon foil to a sufficiently high negative potential. In the course of the measurements, however, it was realized that the LSP allows one to determine not only the polarization of protons or deuterons but also the nuclear polarization of the H_2^+ or D_2^+ ions and, thus, deduce the nuclear polarization of the molecules in the cell gas.

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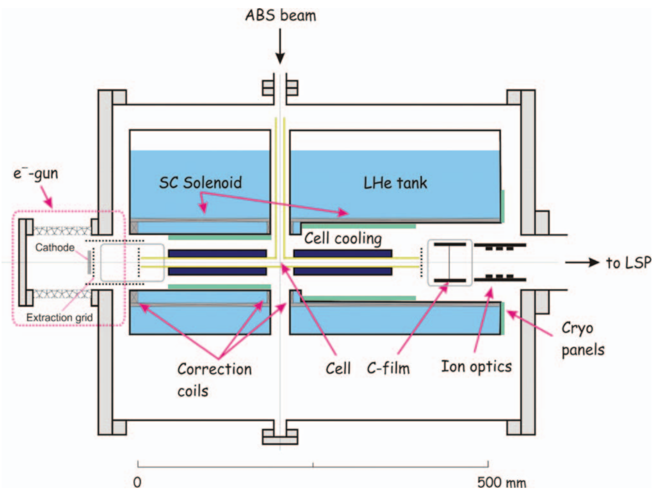

FIG. 1. ISTC setup[10] with the storage cell fed by the polarized atomic beam from the atomic beam source (ABS), with the superconducting (SC) solenoid inside a liquid helium tank, the heating jackets around the cell (full black) for cell-temperature regulation, and the cryogenic pumping panels (green). From left to right, one can see the electron gun, the cell, and the electrodes to accelerate and focus the extracted ions into the LSP. The carbon film is installed whenever dissociation of the extracted molecular ions is desired.

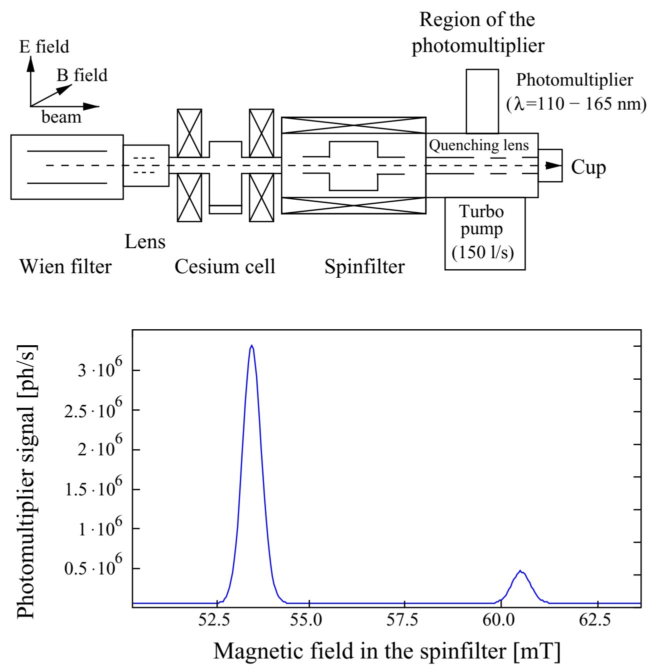

FIG. 2. Schematic drawing of the LSP components with the ion beam coming from the left. In the Wien filter, the electric and magnetic field directions are perpendicular mutually and to the beam direction. The coils at the Cs cell and the solenoid around the spinfilter produce the longitudinal magnetic field along the beam axis.

This novel feature of the LSP is discussed in Sec. III. The results of modeling calculations are presented in Sec. IV. Conclusions and an outlook are found in Sec. V.

II. COMPONENTS OF THE LAMB-SHIFT POLARIMETER

The schematic setup of the Lamb-shift polarimeter is shown in Fig. 2. Here, only the essential features of the components are discussed, and for a more detailed description the reader is referred to Ref. 3.

- The **Wien filter**, the entrance component of the LSP, with its crossed magnetic and electric fields can be used as a mass filter to allow either only protons or only H_2^+ ions to reach the cesium cell due to their different velocities after the acceleration to the same energy behind the storage cell. Due to the longitudinal magnetic field in the storage cell, the protons and H_2^+ ions enter the Wien filter with the polarization vector along the beam direction. In addition, the Wien filter can be used to rotate the polarization vector of the proton[13] by a well defined angle in the plane perpendicular to the direction of the magnetic field contrary to that of the single-electron ions as discussed in Sec. III A.
- In the **cesium cell**, metastable hydrogen atoms are produced by charge exchange of the protons (or deuterons) with the Cs vapor.[14] For H_2^+ ions, this process was mentioned as being observable.[15] A strong longitudinal magnetic field of about 50 mT is applied to preserve the nuclear polarization. For protons and H_2^+ ions, the charge-exchange is discussed in Secs. III B and III C.
- The **spinfilter**[16] is used to transmit at a selected magnetic field strength exclusively metastable atoms in a single hyperfine substate and to quench all other states into the ground state. At a magnetic field of 53.5 mT the metastable H atoms in the hyperfine substate $\alpha 1 := |m_J = +1/2, m_I = +1/2\rangle$ are transmitted, and at 60.5 mT those in the substate $\alpha 2 := |m_J = +1/2, m_I = -1/2\rangle$. Here m_J and m_I denote the magnetic quantum numbers of electron and proton, respectively.
- In the **quench chamber**, the transmitted metastable atoms are quenched into the ground state by a strong electric field of an electric lens due to the Stark effect. The emitted Ly-α photons are registered with a photomultiplier as a function of the magnetic field strength in the spinfilter. The ratio of the photons counted from atoms, transmitted by the spinfilter at the two magnetic field strengths, 53.5 mT for protons in the $m_I = +1/2$ state and 60.5 mT for those in the $m_I = -1/2$ state, yields the polarization of the protons in the beam of metastable $H(2S_{1/2})$ atoms behind the cesium cell. Lyman-α spectra, measured with $H(2S_{1/2})$ atoms from protons, H_2^+ ions, and neutral atoms or molecules incident on the Cs vapor, are discussed in Secs. III B, III C, and III D, respectively.

III. PHYSICAL FEATURES OF THE LSP COMPONENTS

A. Behavior of protons and H_2^+ in the Wien filter

With the crossed electric and magnetic fields, the Wien filter is used as a mass separator for protons (deuterons) and H_2^+ (D_2^+) ions. Transmission for one type of particles is achieved with balanced Coulomb and Lorentz

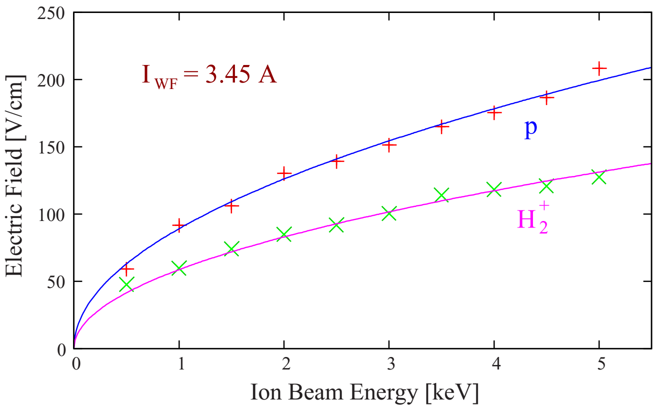

FIG. 3. Electric field strength between the Wien-filter plates, to be applied for transmission of protons or H_2^+ ions at a fixed magnetic field strength of $B_{WF} = 20$ mT, produced by a current of $I_{WF} = 3.45$ A in the Wien-filter coils according to the calibration function $B_{WF}(\text{mT}) = 2.1 + 5.1 \cdot \text{I}_{\text{WF}}(\text{A})$.

force, $\vec{F}_{Coulomb} = -\vec{F}_{Lorentz}$ or $q\vec{E}_{WF} = -q(\vec{v} \times \vec{B}_{WF})$. For non-relativistic energies, the relation between the electric field strength E_{WF} and the magnetic field strength B_{WF} for particles of equal kinetic energy T reads (numerically)

$$E_{WF}(\text{V/cm}) = 4.40 \cdot B_{WF}(\text{mT}) \cdot \sqrt{\frac{T(\text{keV})}{m(\text{u})}}. \tag{1}$$

For 1 keV protons and H_2^+ ions, Fig. 3 shows the agreement of the electric field strengths, applied to reach maximum transmission with B_{WF} set to 20 mT, with the square-root-of-energy dependence of Eq. (1). The figure shows that at a proton and ion beam energy of 1 keV and $B_{WF} = 20$ mT a complete mass separation of protons and H_2^+ ions is obtained with an electric field strength of 87 V/cm for the protons and 61 V/cm for the H_2^+ ions.

For protons of energy E_p and velocity v_p, the dwelling time τ_p in the magnetic field area of the Wien filter of effective length $l = 26.0$ cm is $\tau_p = l/v_p$. For $v_p = 4.4 \times 10^7$ cm/s ($E_p = 1$ keV), τ_p is 5.9×10^{-7} s. From the Larmor frequency ω_p^L of the proton in a magnetic field of strength B_{WF}, the precession angle of the polarization vector during τ_p is

$$\beta_p^L = \omega_p^{\text{L}} \cdot \tau_p = \left(\frac{g_p \cdot \mu_n \cdot B_{WF}}{\hbar}\right) \cdot \tau_p. \tag{2}$$

With the g-factor of the proton $g_p = 5.586$, the nuclear magneton $\mu_n = 3.152 \times 10^{-8}$ eV/T, $1/\hbar = 1.519 \times 10^{15}\ eV^{-1}\text{s}^{-1}$, and $\tau_p = 5.9 \times 10^{-7}$ s one obtains $\beta_p^{\text{L}}(rad) = 1.581 \times 10^2 \cdot B_{WF}$ (T) rad. A complete spin flip of the polarization vector requires a magnetic field strength $B_{WF} = 19.87$ mT. By the spinfilter the projection of the proton polarization vector to the spinfilter axis only is determined. Thus, one expects a cosine dependence for the measured polarization as a function of B_{WF} or the current I_{WF} in the Wien filter coil in agreement with the measured dependence shown in the upper panel of Fig. 4. It is obvious that the maximum value, expected at $I_{WF} = 0$, is measured with an offset of -0.43 A. This is explained by the magnetic hysteresis of the steel in the Wien filter magnet. After each single measurement, the current was increased to a value above 10 A, where saturation starts, and then reduced to the value aimed at. According to this calibration curve of the Wien filter magnet, a current of 0 A corresponds to $B_{WF} = 2.1$ mT or to a rotation of the polarization vector by 19°. The current difference of 3.88 A from the offset at -0.43 A to the minimum at 3.45 A within the errors agrees with the expected value from the linear calibration of the Wien filter below 5 A:

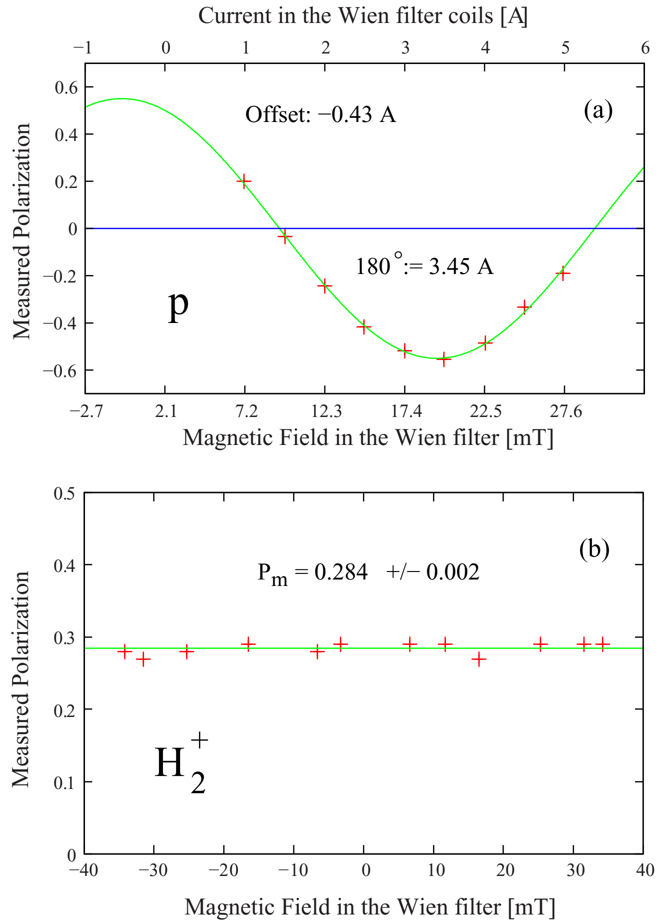

FIG. 4. Upper panel (a): Projection of the polarization vector of 1 keV protons to the beam axis, measured as a function of the magnetic field in the Wien filter, and the fit by a cosine function. Lower panel (b): Measured projection of the polarization vector of the two protons in 1 keV H_2^+ ions showing no influence at all of the magnetic field strength in the Wien filter, even after inversion of the field direction.

$$B_{WF}(mT) = 2.1 + 5.1 \cdot I_{WF}(A). \tag{3}$$

A completely different dependence on the magnetic field in the Wien-filter coil is observed for H_2^+ ions. The lower panel of Fig. 4 shows the proton polarization deduced from the Lyman-α radiation of the metastable H atoms created in the interaction of the H_2^+ ions with the vapor in the cesium cell. Here, the measured projection of the polarization vector to the beam axis does not depend at all on the magnetic field strength in the Wien filter. Even reversal of the magnetic field direction does not influence the measured polarization. The difference to the protons is caused by the electron in the H_2^+ ion which dominates the magnetic moment of the ion. With $g_e = 2$ and the Bohr magneton $\mu_{\text{B}} = 0.579 \times 10^{-4}$ eV/T its Larmor frequency ω_e^L is by about three orders of magnitude larger than that of a proton. Therefore, the magnetic moment of the electron follows the magnetic field direction adiabatically. Within the range of the magnetic field in the Wien filter, the magnetic moment of the electron is directed along the magnetic field and perpendicular to the beam direction. It follows the transient field between the Wien filter and the longitudinal field in the cesium cell. The nuclear magnetic moments of the protons follow those of the electrons, because their coupling to the magnetic field, produced by the spins of the electrons on the order of 10 T, is by orders of magnitude stronger than the external magnetic field of the Wien filter. Thus, the polarization of the protons in the H_2^+ ions in the longitudinal magnetic field in the cesium cell and in the subsequent spinfilter is identical to that in the storage cell, independent of the magnetic field strength in the Wien filter.

B. Charge exchange of polarized protons with Cs atoms in the cesium cell

The production of metastable hydrogen $H(2S_{1/2})$ atoms by charge exchange of protons with Cs atoms is well known and was exploited in a variety of applications. The yield of $H(2S_{1/2})$ atoms with 1 keV protons peaks at an areal number density of about 1.3×10^{14} Cs atoms per cm^2.[14] With the present cell setup and the magnetic holding field strength of 50 mT, the maximum yield was found for a temperature of $T_b = 135\,^\circ$C of the liquid Cs at the bottom of the cell.[17] Nevertheless, most of the measurements described here are made at temperatures between 150 and 160°C.

The polarization $P_{Ly}(B)$, determined behind the charge exchange in the Cs cell from the Lyman-α spectrum, for protons of initial polarization P_i has been shown to be[3]

$$P_{Ly}(B) = \frac{P_i \cdot [1 + a(B)]}{2 + P_i \cdot [1 - a(B)]}. \quad (4)$$

Here $a(B) = (B/B_c) \cdot [1 + (B/B_c)^2]^{-1/2}$, where $B_c = 6.34$ mT is the critical magnetic field strength of the metastable hydrogen atoms. The maximum current in the coils around the Cs cell of 20 A corresponds to a magnetic field strength $B = 50$ mT. At the given values of B_c and B according to Eq. (4) 99.4% of the initial proton polarization is preserved in the metastable atoms. A typical Lyman-α spectrum is shown in Fig. 5. The storage-cell gas was delivered by the polarized atomic beam source with H atoms of positive vector polarization of about +0.89.[11] These atoms recombine on the gold surface inside the storage cell into molecules. The protons from the dissociated molecules were accelerated to 1 keV. In the Wien filter the polarization vector was reversed. From the spectrum as shown in Fig. 5, a proton polarization of -0.46 ± 0.005 and thus (due to the reversal in the Wien filter) a value of $+0.46 \pm 0.005$ was deduced. Due to the strong recombination of the atoms into molecules it was measured that only 0.7% of the particles inside the storage cell are H atoms that contribute 3.8% of the protons with a nuclear polarization of $P_p = 0.89 \pm 0.01$. (Ionization cross section at a kinetic energy of the electrons of 150 eV: $\sigma_{H->p} = 4.6 \times 10^{-17}$ cm^2/$\sigma_{H_2->p} = 0.8 \times 10^{-17}$ cm^2 [18]). Therefore, the polarization of the H_2 molecules is $P_M = 0.443 \pm 0.006$.

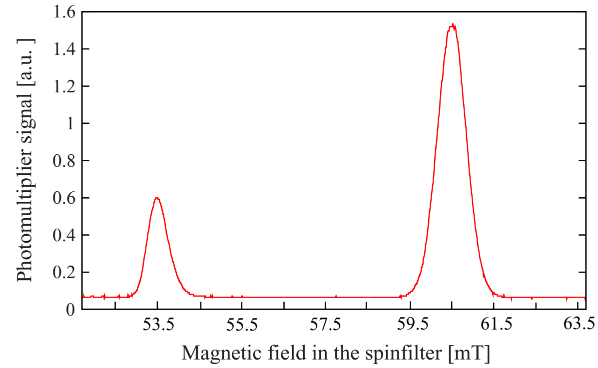

FIG. 5. Lyman-α spectrum from metastable hydrogen atoms originating from 1 keV protons incident on the Cs vapor (temperature of the liquid Cs $T_b = 150\,^\circ$C, magnetic field strength in the Cs cell $B_{Cs} = 50$ mT). Neglecting small corrections,[3] the nuclear polarization P_{Ly} of the metastable atoms is obtained from the relative ratio of the peak heights as $P_{Ly} = -0.46 \pm 0.005$.

C. Interaction of polarized H_2^+ ions with Cs atoms in the cesium cell

The possibility to produce metastable hydrogen atoms by the interaction of H_2^+ ions with Cs atoms was mentioned before.[15] The present setup allowed us to study the efficiency of this process relative to the charge exchange with protons. The Wien filter was used to separate the proton and H_2^+ beams and the magnetic field strength in the Cs cell was kept at 50 mT. Without Cs vapor in the cell, proton and H_2^+ beams of varying intensities were extracted from the storage cell and the beam intensities were measured with the Faraday cup indicated in Fig. 2. Each of these measurements was followed by a measurement of the Lyman-α intensity with the liquid Cs heated to $T_b = 150\,^\circ$C. As an average value the intensity ratios show that the efficiency of producing metastable hydrogen atoms in the interaction of H_2^+ ions with Cs atoms is (2.9 ± 0.4)% of that of producing them from protons.

A typical Lyman-α spectrum, emitted by metastable hydrogen from the interaction of H_2^+ ions with the Cs vapor as a function of the magnetic field strength in the spinfilter, is presented in Fig. 6.

The spectrum of Fig. 6, measured with the H_2^+ beam, demonstrates that nuclear polarization is maintained (i) in the recombination process and (ii) in the interaction with the Cs atoms leading to metastable atoms. In this case, the polarized hydrogen atoms recombine on a water surface cooled down to 50 K into H_2 molecules. The nuclear polarization of the H_2 molecules is now measured directly as $P_M = 0.31 \pm 0.01$ and smaller compared to the $P_M = 0.443 \pm 0.006$ measured for the recombination on a gold surface before.

An evident difference between the spectrum of Fig. 6, measured with the H_2^+ beam, and that of Fig. 5, measured with the proton beam, is the peak-to-background ratio. In both cases, the background intensity of the peak regions does not depend on the magnetic field strength in the spinfilter. It is understood as resulting from excitation processes in the

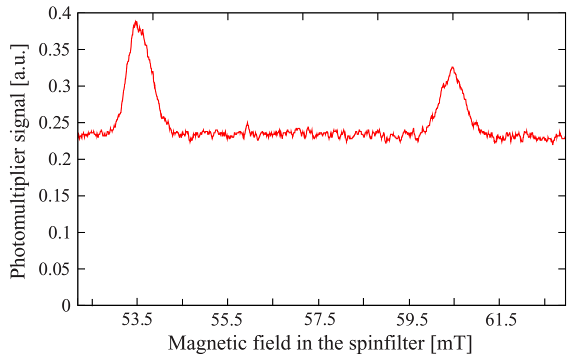

FIG. 6. Lyman-α spectrum from metastable hydrogen atoms originating from H_2^+ ions incident on the Cs vapor ($T_b = 150\,^\circ$C, $B_{Cs} = 50$ mT). The relative ratio of the peak heights yields the nuclear polarization of the metastable atoms of $+0.31 \pm 0.01$. The absolute yield of both peaks is (2.9 ± 0.4)% of those in Fig. 5.

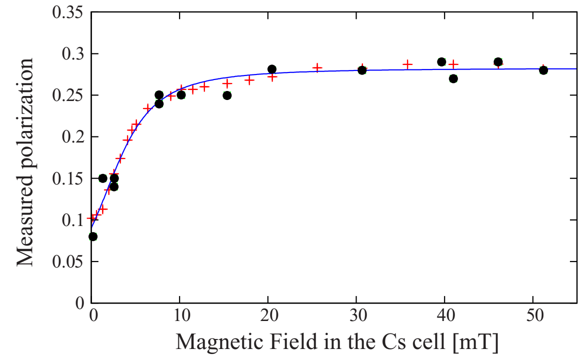

FIG. 8. Polarization P_{Ly} of the H_2^+ ion beam, measured as a function of the magnetic field in the cesium cell (black •), compared with the dependence predicted by Eq. (4) with $B_c = 6.34$ mT (blue line) and with the proton-beam data (red +) taken from Ref. 3 and normalized to the present data by the saturation values.

interaction of the neutral beam particles, transmitted by the spinfilter, and the residual gas in the quench chamber.[17] For equal proton and H_2^+ beam intensities, incident on the Cs vapor, the intensity of transmitted atoms is higher by a factor of two for the incoming H_2^+ beam due to a stripping process of the H_2^+ into H^+ at the Cs cell (see Sec. V).

As described before, the cesium temperature for the maximum yield had been found for protons incident on the Cs vapor at 136 °C.[17] The result of a corresponding measurement with 1 keV H_2^+ ions incident on the Cs vapor is shown in Fig. 7. As for the proton, the maximum value is measured for T_b around 140 °C, which indicates similar processes in the Cs vapor.

In Sec. III B, it was discussed how the polarization P_{Ly} of a proton beam, deduced from the peak ratio in the Lyman-α spectrum, depends on the magnetic field strength in the Cs cell. The correctness of Eq. (4) was confirmed by an earlier measurement.[3] A corresponding measurement was performed with a beam of H_2^+ ions. In Fig. 8, the measured B-field dependence is compared with the prediction of Eq. (4) and with the normalized values measured with a proton beam. Both sets of experimental data in excellent agreement confirm the prediction calculated with $B_c = 6.34$ mT. In the proton case,

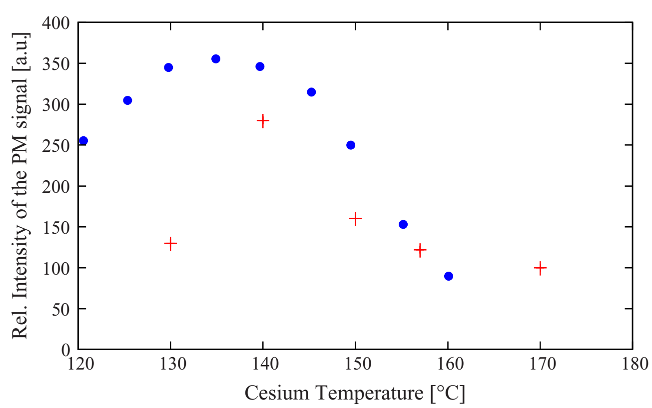

FIG. 7. Average peak intensity of the Lyman-α spectra, taken with 1 keV H_2^+ ions (red +) or protons (blue •, taken from Ref. 17) incident on the Cs vapor, as a function of the temperature of the liquid Cs in the Cs cell. (Magnetic field strength in the Cs cell kept fixed at 50 mT.)

the B dependence gives the probability that the proton polarization is preserved in the charge exchange with a Cs atom and creation of a metastable $H(2S_{1/2})$ atom. The agreement of both experimental B dependences shows that in the interaction of the H_2^+ ions in the Cs vapor the applied B field of 50 mT guarantees the preservation of the nuclear polarization of the two protons within 99%. Therefore, only neutral metastable atoms could have been produced during the charge exchange with the Cs atoms. If hydrogen atoms in the ground state (1S) would be produced during an intermediate step of the metastable production, e.g., dissociative neutralization with the Cs vapor, the observed critical field must be $B_c = 50.7$ mT.

D. Transmission of neutral molecules and atoms by the Wien filter

For the measurements with protons and H_2^+ ions, the electric field strength in the Wien filter was set for transmission of $u = 1$ and $u = 2$ according to Eq. (1). To study a possible leakage through the Wien filter, the electric field strength was set for transmission of particles with $u = 0.5$, 1.5, and for very large masses. For the recombination on fused quartz glass the measured unexpected Lyman-α spectrum, observed for the first time, is shown in Fig. 9. The observed polarization of $+0.36 \pm 0.01$ lies between those resulting with transmission of protons ($P_p = 0.43 \pm 0.01$) produced from atoms and molecules and H_2^+ ions ($P_M = 0.23 \pm 0.01$). The measurement of the dependence of the average peak intensity on the temperature of the liquid Cs in the cesium cell yields the maximum value around $T_b = 135\,^\circ$C as found for the protons and H_2^+ ions. An explanation of this effect is charge exchange of the accelerated H_2^+ and protons ions in the gas between the storage cell and the Wien filter. The neutral H_2 molecules and H atoms then pass the Wien filter without being affected by the electric and magnetic fields. In the Cs vapor they encounter the processes leading to the production of metastable $H(2S_{1/2})$ atoms.

This effect, responsible for a small correction in the calculation of the polarization from the Lyman-α spectrum, was not seen before, when the atomic hydrogen beam was ionized in an electron-impact ionizer[3,4] and separated from the gas

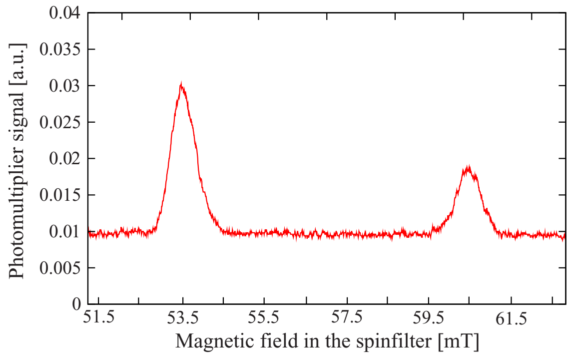

FIG. 9. Lyman-α spectrum from metastable hydrogen atoms produced in the Cs vapor with the Wien filter set to transmission of particles with mass "$u = 1.5$", i.e., neither protons nor H_2^+ ions ($T_b = 150\,°$C, $B_{Cs} = 50$ mT). The relative ratio of the peak heights yields a nuclear polarization of the metastable atoms of $+0.36 \pm 0.01$. The absolute average yield of both peaks is on the order of 1% of those in Fig. 6.

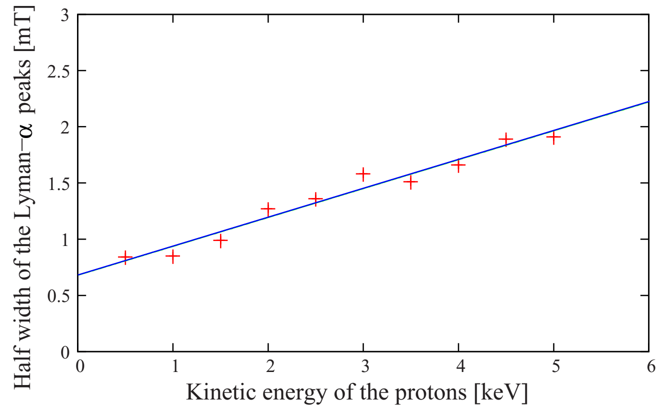

FIG. 10. Measured width ΔB_{SF} (FWHM) of the peaks in the Lyman-α spectrum as a function of the kinetic energy T_A of the hydrogen atoms in the spinfilter, defined by the kinetic energy of the protons at the entrance of the Wien filter. The linear fit function is ΔB (mT) $= 0.67 + 0.26 \cdot T_A$ (keV) with a relative uncertainty of 0.07 (rf power ~80 mW, $E_{static} \sim 7$ V/cm).

jet directly behind the ionizer. Just the residual gas at a pressure of 10^{-6} mbar on a path length of about 20 cm in front of the Wien filter could not produce enough neutral particles to contribute significantly. During these measurements, the path length of the protons and H_2^+ ions with about 45 cm was twice longer, but the vacuum pressure in the ISTC chamber was with $\sim 10^{-7}$ mbar much lower than before. Our explanation for the now observed neutralization of the accelerated protons and H_2^+ ions is the increased gas density due to the flux of the hydrogen atoms and molecules leaving the cell along the ion beam trajectories. But due to the unknown cross section for the production of metastable hydrogen atoms during the collisions of fast (1 keV) atoms and molecules with cesium vapor a detailed calculation is not possible.

E. The peak widths in the Lyman-α spectra measured with protons and H_2^+ ions

By the spinfilter, the hydrogen atoms in the metastable $2S_{1/2}$ states with one of the nuclear quantum numbers $m_I = +1/2$ or $m_I = -1/2$ are transmitted and those in the states with the other one are quenched to the ground state. This separation is due to the "three-level interaction"[19] of the atoms in magnetic and electric fields and an rf electric field. The rf field induces oscillations between the two hyperfine (HF) states of the metastable $2S_{1/2}$ state ("α" branch, $\tau = 0.14$ s, electron quantum number $m_J = +1/2$) and the degenerate HF states of the "β" branch of the metastable $2S_{1/2}$ state ($m_J = -1/2$) and the "e" branch of the short-lived $2P_{1/2}$ state ($\tau = 10^{-9}$ s, $m_J = +1/2$). The energy uncertainty ΔE_{tr} of the transitions between the HF states with $m_{I=+1/2}$ and of those with $m_{I=-1/2}$ depends on the rf power and the electric field and decreases with increasing dwelling time t of the atoms in the field region of the spin filter. Because t is proportional to the inverse of the atom velocity v_A, ΔE_{tr} increases with increasing velocity. A quantum-mechanical treatment yields a linear dependence on v_A^2,[20] i.e., a linear dependence of ΔE_{tr} on the kinetic energy T_A. When the rf power and the electric field are constant this dependence is confirmed by the data of Fig. 10, where ΔE_{tr} is expressed by the measured resonance width of the ramped magnetic field. The width ΔB_{SF}, expected for the peaks with beams of 1 keV protons, H_2^+ ions, and transmitted H_2 molecules according to the relation given in the caption of Fig. 10, can be compared with the measured widths of Figs. 5, 6, and 9, respectively. As expected, the width ΔB_{SF} of the proton measurement is broader by about 15% compared to the H_2^+ ions and the neutral particle measurements for stable rf power and static electric field.

IV. MODEL CALCULATIONS

The possibility to separate the accelerated protons and the H_2^+ ions in the beam from the gas-storage cell with the use of the Wien filter was discussed in Sec. III A. The possibility to measure the nuclear polarization of H_2^+ ions was confirmed by the present experiments. The yield of metastable hydrogen atoms resulting from H_2^+ ions, incident on Cs vapor, is described using the appropriate cross sections in the interaction processes. The validity of the formalism used is investigated by its application to describe measured yields for protons and deuterons incident on Cs vapor.[14,17] In addition, it is used to describe the yield of metastable hydrogen atoms in the $2S_{1/2}$ state, measured for 1 keV H_2^+ ions as function of the temperature at the bottom of the Cs cell in the range $T_b = 110$ to 160°C (Fig. 7), and to understand the ratio of yields for 1 keV H_2^+ ions and protons, measured as (2.9 ± 0.4)% at $T_b = 135$°C (Sec. III C).

A. The modeling function for protons or deuterons incident on Cs vapor

The yield $f_m(p)$ or $f_m(d)$ of atoms, leaving the vapor in the Cs cell in the metastable $2S_{1/2}$ state, results from a two-step process. Charge exchange with the Cs atoms with a cross section σ_{+m} leads to the population of the metastable $2S_{1/2}$ state, whereas the ground state and states with prompt radiative decay to the ground state are populated with a cross section σ_{+r}. Subsequently, atoms get lost from the $2S_{1/2}$ state by collisional destruction in the Cs vapor with a cross section σ_{cd}. The labeling of the cross sections follows that of Ref. 14. In analogy to the formalism of nuclear decay chains (e.g., Ref. 21) a method was developed to calculate the yields $f_m(p)$ or $f_m(d)$ with time replaced by the areal number density n of the Cs vapor and the decay constants by the cross sections,

$$f_m(x) = I[X(S_{1/2})]/I(x) = \frac{\sigma_{+m}}{\sigma_{+m} + \sigma_{+r}} \frac{\sigma_{tot}}{\sigma_{cd} - \sigma_{tot}} [e^{-\sigma_{tot} n} - e^{-\sigma_{cd} n}]. \quad (5)$$

The x stands for the incident particle type, $I[(X(S_{1/2}))]$ for the intensity of metastable hydrogen ($X = H$) or deuterium ($X = D$) atoms behind the Cs vapor, and $I(x)$ for the intensity of the beam incident on the Cs vapor. The first fraction takes care of the branching ratio in the charge-exchange process. The other two terms containing $\sigma_{tot} = \sigma_{+m} + \sigma_{+r}$ describe the reaction chain. The cross sections depend on the type and the energy of the incident particles.

B. Application to deuterons incident on Cs vapor

The fractional yield $F_m(d)$ relative to the total outgoing beam for 1 keV deuterons has been measured as a function of n up to 1.2×10^{15} Cs atoms/cm^2.[14] Different from the definition of f_m, the fractional yield F_m is defined as $I_m/(I_m + I_g + I_- + I_+)$, where I_m, I_g, I_-, and I_+ are the intensities of metastable atoms, ground-state atoms, negative ions, and residual deuterons, respectively, in the outgoing beam. By definition $F_m + F_g + F_- + F_+ = 1$. To describe the measured fractional yield by the n dependence of Eq. (5), the cross sections for 1 keV deuterons should have values near those measured for 0.5 keV protons as $\sigma_{+m} = 6.0 \times 10^{-15}$ cm^2, $\sigma_{+r} = 3.8 \times 10^{-15}$ cm^2, and $\sigma_{cd} \sim 5.0 \times 10^{-15}$ cm^2.[14] In Fig. 11, the calculated n dependences are compared with the measured data.

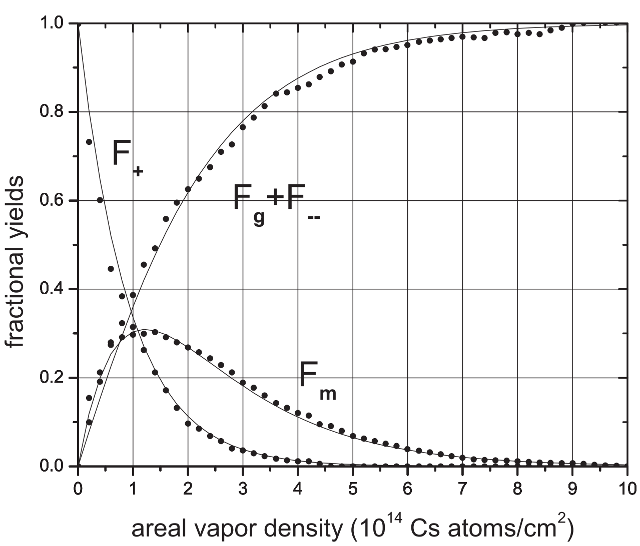

FIG. 11. Calculated (full lines) and measured (dots, taken from Fig. 11 of Ref. 14) fractional yields in the beam behind Cs vapor for incident 1 keV deuterons as a function of the areal vapor density n. F_m: metastable $D(2S_{1/2})$ atoms, F_+: residual deuterons, $F_g + F_-$: ground-state atoms and D^- ions.

The fractional yield $F_+(n)$ is calculated using the n dependence $1-\exp(-\sigma_{tot}n)$. The measured fractional yields F_g and F_- are combined in one data set, the calculated n dependence is $1 - F_m - F_+$. Satisfactory agreement is achieved with $\sigma_{+m} = 7.0 \times 10^{-15}$ cm^2, $\sigma_{+r} = 3.9 \times 10^{-15}$ cm^2, and $\sigma_{cd} = 6.0 \times 10^{-15}$cm^2, all close to the values given above. The agreement demonstrates that the relation of Eq. (5) is adequate to describe the two-step process in the Cs vapor.

C. Application to protons incident on Cs vapor

For a series of incident proton energies between 0.5 and 2.5 keV, the absolute fractional yields F_m have been measured as a function of the areal number density n of the Cs vapor.[14] Figure 12 shows the data for 0.9 keV (dots) with the maximum at $n = 1.5 \times 10^{14}$ Cs atoms/cm^2 and the n dependence calculated with Eq. (5). The used cross sections $\sigma_{+m} = 3.1 \times 10^{-15}$ cm^2, $\sigma_{+r} = 5.6 \times 10^{-15}$ cm^2, and $\sigma_{cd} = 6.5 \times 10^{-15}$ cm^2 are close to the measured values $\sigma_{+m} = 2.8 \times 10^{-15}$ cm^2, $\sigma_{+r} = 6.1 \times 10^{-15}$ cm^2, and $\sigma_{cd} \sim 5 \times 10^{-15}$ cm^2.[14] Both, the measured and the calculated n dependences have the maximum at $n \approx 1.5 \times 10^{14}$ Cs atoms/cm^2.

In an earlier work,[17] relative production rates of metastable $H(2S_{1/2})$ atoms by 1 keV protons, incident on Cs vapor, had been measured at temperatures T_b of the bottom of the Cs cell in the range from 110 to 160 °C. The production rates, measured with constant incident proton intensity, were given by the current in the photomultiplier at the quench chamber as a function of T_b. For comparison with the measured and calculated fractional yield, the temperature has to be converted to the areal number density of the Cs vapor. According to the Taylor-Langmuir curve,[14,22] the saturated vapor pressure of Cs as a function of the temperature for the liquid

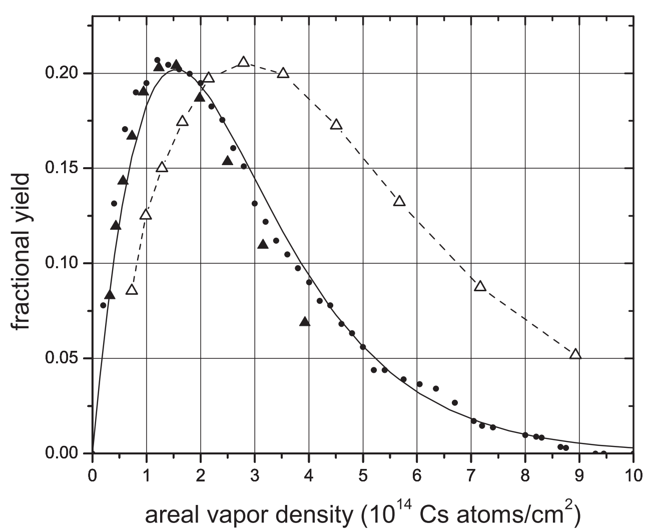

FIG. 12. Calculated (full line) and measured (dots, taken from Fig. 12 of Ref. 14) fractional yield F_m in the beam behind Cs vapor for incident 0.9 keV protons as a function of the areal vapor density n. Open triangles: measured relative values of F_m with 1 keV protons (Fig. 3.9 of Ref. 17) as a function of the areal vapor densities corresponding to the saturated Cs vapor pressure. Full triangles: the same measured relative values for areal vapor densities reduced by a factor of 0.44. In both cases, the maximum of the data set is normalized to the maximum value of the dots and the full line.

phase ($T_b > 302$ K) is

$$\log_{10} p\,(\mathrm{Torr}) = 11.0531 - 1.35 \cdot \log_{10} T_b\,(\mathrm{K}) - \frac{4041}{T_b\,(\mathrm{K})}. \tag{6}$$

Under the assumption that the vapor contains atomic Cs only and that it can be regarded as an ideal gas, the Cs number density n', given in units of Cs atoms/cm^3 in the vapor, is

$$n'(p, T) = n'_n \cdot \frac{p}{p_n} \cdot \frac{T_n}{T}, \tag{7}$$

where $n'_n = 2.69 \times 10^{19}$ Cs atoms/cm^3 is the normal number density for $p_n = 1013$ mbar and $T_n = 273$ K. The length of the Cs cell along the proton beam defines the interaction length in the Cs vapor as $l = 2.75$ cm, resulting in an areal number density of the Cs vapor of $n = n' \cdot l$.

The measured values of F_m, attributed to the areal vapor densities corresponding to the saturated vapor pressure, in Fig. 12 are given as open triangles. The maximum, measured for $T_b = 135\,^\circ$C, lies at $n_{max} = 2.8 \times 10^{14}$ Cs atoms/cm^2. Obviously n_{max} is appreciably higher than that of the data shown in the figure for known areal densities and of all data measured for proton energies between 0.5 and 2.5 keV.[14] Thus, one has to conclude that the average vapor pressure in the Cs cell is lower than that of saturated vapor. As is shown in the figure, good agreement with the measured and calculated data is obtained, when the Cs vapor pressure and thereby the areal number density is reduced by a factor 0.44. This fits with the fact that the wall of the Cs cell around the beam line is colder than the bottom of the cell leading to a reduced vapor density. This result is essential for the analysis of the production rates of metastable $H(2S_{1/2})$ from H_2^+ ions incident on the Cs vapor in the same Cs cell.

D. Modeling of the yield of metastable *H* atoms from H_2^+ ions incident on Cs vapor

The total charge-exchange and breakup cross section for 3 to 23 keV D_2^+ incident on Cs vapor has been measured as $(1.36 \pm 0.08) \times 10^{-14}$ cm^2 at 3.0 keV.[23] The measurements show that the reactions $D_2^+ + Cs \rightarrow D_2^0 + Cs^+$ and $D_2^+ + Cs \rightarrow 2D^0 + Cs^+$ dominate. The aim of this experiment like that of other studies,[24,25] and the given references, was to determine the fraction of D^- ions in the beam behind the Cs vapor for ion sources. Therefore, no attempts were made to separate the fractions of D^0 and D_2^0 created in the initial charge-exchange reaction. The cross sections of ions, atoms and molecules of deuterium can be used for those of hydrogen of identical velocity, i.e., of half the kinetic energy as discussed by Ref. 24. The excellent agreement of the polarization measured with incident protons and H_2^+ ions and the calculation with the critical magnetic field strength $B_c = 6.34$ mT of the metastable hydrogen atom (Fig. 8) leads to the conclusion that for the H_2^+ ions as for the protons the preservation of the polarization of the incident particles is to be attributed to the creation of metastable $H(2S)$ atoms in the charge-exchange process under the magnetic field strength of 50 mT applied to the Cs cell. Conflicting results were measured concerning the production of the atoms in excited states

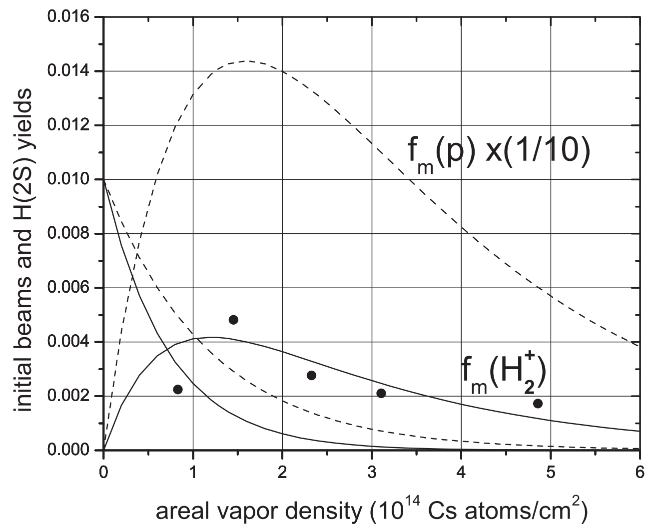

FIG. 13. Calculated yields $f_m(H_2^+)$ (full line) and $f_m(p)$ (dashed line) of metastable $H(2S)$ atoms for 1 keV H_2^+ ions and protons, incident on Cs vapor as a function of the areal vapor density n, and the measured relative intensity of the Lyman-α peaks in Fig. 7 (full dots, normalized to the calculated curve for $f_m(H_2^+)$ and attributed to the reduced areal Cs vapor densities). The exponentials show the decrease of the incident beams in the Cs vapor (full line: H_2^+; dashed line: protons).

in dissociative collisions of H_2^+ ions of kinetic energy ≥ 1 keV incident on H_2.[26] Other measurements of this author with 10 keV H_2^+ ions on H_2, He, and Ar targets indicate that electron capture into the repulsive $b^3\Sigma_u^+$ state in H_2 plays a dominant role.

In the present modeling calculation, the assumption was made that a small fraction of the total electron-capture cross section $\sigma_{tot} \approx 1.36 \times 10^{-14}$ cm^2 for 1 keV H_2^+ ions incident on Cs[24] is attributed to the direct creation of metastable $H(2S)$ atoms. Accordingly in Eq. (5) $\sigma_{+r} = \sigma_{tot} - \sigma_{+m}$, where $\sigma_{+m} \ll \sigma_{tot}$. A factor of 2 has to be included to take into account the breakup of the molecular ion. Furthermore, the ratio $f_m(H_2^+)/f_m(p) = (2.9 \pm 0.4)\%$ has to be reproduced, which was measured at $T_b = 135\,^\circ$C, i.e., at the reduced areal vapor density of $n = 1.23 \times 10^{14}$ Cs atoms/cm^2. The result is shown in Fig. 13. The n dependence of $f_m(H_2^+)$ yields $\sigma_{tot} = 1.36 \times 10^{-14}$ cm^2 and $\sigma_{+m} = 5.0 \times 10^{-17}$ cm^2, i.e., $\sigma_{+m}/\sigma_{tot} = 3.6 \times 10^{-3}$. Further, $\sigma_{tot} = 6.5 \times 10^{-15}$ cm^2 has the value used above. The two exponential curves show the decrease of the incident proton and H_2^+ beam intensity in the Cs vapor. Due to the higher value of σ_{tot} for H_2^+, 1.36×10^{-14} cm^2 compared to 8.6×10^{-15} cm^2, the decrease for H_2^+ is steeper, leading to a negative shift of the maximum of the H_2^+ curve against that for the protons. The calculated curve for H_2^+ reproduces the general dependence of the measured signal intensities of the Lyman-alpha peeks due to the amount of metastable atoms on the areal Cs vapor density.

V. CONCLUSIONS AND OUTLOOK

The results of the present measurements show that the standard components of a Lamb-shift polarimeter—Wien filter, Cs vapor cell, spinfilter, and quench chamber—allow to separate the H_2^+ ions from the protons in 1 keV beams and to measure the nuclear polarization of both components with a precision of 2% or better. The separation of the two beam components is achieved with the Wien filter by setting the magnetic field strength to a value that corresponds to an orientation of the polarization vector parallel or antiparallel to the beam direction, and setting the electric field strength to a value that fulfills the transmission relation for either the protons or the H_2^+ ions (see Eq. (1) and Fig. 3). Due to the high Larmor frequency, the polarization vector of the single-electron ions follows adiabatically the magnetic field direction and thus the polarization along the beam direction is conserved. In addition, the nuclear polarization of fast H_2 molecules (∼1 keV) is measurable due to the fact that the molecules are transformed partly into polarized metastable H atoms by charge exchange in the Cs cell. The performed model calculations reproduce the measured yields of metastable $H(2S)$ atoms in the beam behind the Cs vapor and their dependence on the areal Cs vapor density when using the known interaction cross sections (Fig. 13).

The different polarized ion sources worldwide have shown that the nuclear polarization is not influenced much during ionization of H atoms into protons, charge exchange into H^- ions or stripping of H^- into protons unless a strong magnetic field is attached to decouple the nuclear from the electron spin. Therefore, we expect in first order no polarization losses during the different transformations of the H atoms and H_2 molecules. Several small correction factors like for the finite magnetic field at the Cs cell or during the ionization process are described in Refs. 3 and 4. In principle, only a calibration of the nuclear polarization, e.g., with use of the known analyzing powers of nuclear reactions like in Ref. 7 can show, if polarization is lost during the ionization process and the transportation of the ions through the Lamb-shift polarimeter. But in the meantime, we have found a material that conserves the nuclear polarization of the atoms almost completely in the molecules during the recombination process. On a Fomblin (Perfluorpolyether/PFPE, a product from Solvay Solexis) surface at 100 K the polarization of the H_2^+ ions was measured to be $P_M = -0.84 \pm 0.02$ and the polarization of the protons was $P_p = -0.81 \pm 0.02$. The storage cell was fed with atoms in the hyperfine state 3 with a polarization of $P = -0.94 \pm 0.01$. In Ref. 4 was shown that about $(2.9 \pm 0.4)\%$ of the ABS beam particles are unpolarized H_2 molecules due to a ballistic flux from the ABS nozzle and the different vacuum chambers. Therefore, the upper limit for the nuclear polarization of the molecules is $P_{max} = -0.88 \pm 0.015$. In addition, another small amount of polarization is lost due to the recombination on a water surface that cannot be avoided completely on such cold surfaces. During these measurements the Fomblin surface was rather clean but, nevertheless, at least $\Delta P = 0.02\%$ are lost. In addition, unpolarized H_2 molecules from residual gas can decrease the upper limit of the polarization too. At this time, it should be mentioned that it is possible to produce unpolarized protons from water or other residual gases like carbon hydrides so that the measured polarization of the protons is always slightly smaller than the polarization of the H_2^+ ions. Therefore, we can conclude that the polarization losses during the transformations of the H_2 molecules in the different components of the LSP and their magnetic fields are less than 0.02.

The calculations of the interaction processes in the Cs vapor confirm that the nuclear polarization of both protons and H_2^+ ions with use of the spinfilter and the Lyman-α radiation, emitted in the quench chamber, can be measured with highest efficiency without change of the areal vapor density in the Cs cell, i.e., the temperature of the liquid Cs. The fit of the data as a function of the temperature at the liquid Cs reservoir in the cell shows that the average Cs vapor density along the beam-interaction zone is lower by a factor 0.44 than that of saturated vapor at bottom temperature.

With polarized deuteron beams from the atomic beam source,[11] measurements with beams containing D_2^+ ions and deuterons were performed as well. Although this is not discussed here in detail, the results show that with the present technique—formation of metastable deuterium atoms from deuterons and D_2^+ ions in the Cs vapor—their vector and tensor polarization can be measured with separation of the D_2^+ ions from the deuterons in the Wien filter. To separate D_2^+ ions and deuterons, the electric field strengths had to be set to discriminate particles of mass 4 against those of mass 2.

At present and for the near future, the atomic beam source is installed and is being used at the ANKE spectrometer for experiments with the COSY beam. Afterward, it will be used again to feed the storage cell for the studies of the preservation of nuclear polarization of the atomic and recombined molecular hydrogen and deuterium gas in the cell. Special topics will be the studies of the influence of the thin water layer on the inner cell wall and the desorption process of atoms and molecules from different cell-wall materials. These measurements will make use of the present result that the nuclear polarization of protons (deuterons) and H_2^+ (D_2^+) ions, produced by electron impact in the cell gas, can be measured with use of the mass separation in the Wien filter.

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