Generation of nuclear-spin-polarized protons by resonant ionization of hydrogen atoms

H. Rottke and H. Zacharias
Fakultät für Physik, Universität Bielefeld, D-4800 Bielefeld 1, West Germany
(Received 4 June 1985)

An atomic beam of H atoms was crossed by a pulsed beam of narrow bandwidth ($\Delta\nu \sim 700$ MHz) circularly polarized tunable vuv laser radiation at $\lambda = 121.6$ nm. The atoms were excited into a single magnetic substate in the $2p\ ^2P_{1/2}$ level and thus polarized. This polarization is transferred via hyperfine interaction to the nucleus. A second tunable uv laser served to ionize these atoms, thereby producing nuclear-spin-polarized protons with, theoretically, 100% degree of polarization. By changing the helicity of the circularly polarized vuv laser with a quarter-wave plate, the polarization direction of protons is changed.

Polarized proton beams have proven to be a very powerful tool in nuclear and high-energy physics, as witnessed by the three recent major conferences on this subject.[1-3] Usually the nuclear-spin polarization of the protons is created by electron impact ionization of either polarized ground state[4-6] or magnetically selected polarized metastable 2^2S hydrogen atoms.[5,6] At high energies stripping ionization of polarized H^- beams is frequently used.[1,2] Recently, charge exchange collisions between unpolarized beams and polarized targets, mostly alkali-metal atom vapors, have been developed as polarized ion sources.[7,8] Although these methods are reliable and well established, they have some disadvantages. (1) Depolarization effects through electron impact ionization of the polarized atoms[6] and in the charge exchange collisions between H atoms and polarized alkali-metal atoms[9] limit the degree of proton polarization to about 60–80% in practical applications. (2) A change of the polarization direction may require large magnetic fields and superconducting devices.[10] (3) The ionization cross section of H atoms by electron impact is fairly low. (4) Often other charged particles are created in the last stage of polarized proton production, which makes a mass selective ion extraction necessary. (5) Short pulse proton beams which are desirable to achieve a higher signal-to-noise ratio, are not simple to obtain.[11]

We want to report results of two-step photoionization experiments on hydrogen atoms with high spectral resolution, demonstrating an alternative approach for the efficient production of spin-polarized protons. This method can provide 100% nuclear-spin-polarized protons without generating charged atoms or molecules other than the H^+ ions. Depending on the exciting laser power, ionization efficiencies of greater than 70% are feasible. In addition, a convenient and straightforward means of changing the direction of the nuclear-spin polarization is possible.

In resonantly enhanced two-step photoionization one can make use of the quantum structure of the atomic system for selecting a specific state to be ionized; this is not possible in single-photon ionization or in electron impact ionization. The selection rules for polarized dipole radiation enable one to impose an alignment or an orientation upon the atom prior to ionization. In the case of atomic hydrogen, irradiation with σ^+ polarized light allows the population of the *single* magnetic substate, $m_F = +1$, in the $2p\ ^2P_{1/2}$ fine-structure level, if the initial state is the $(1s\ ^2S_{1/2}, F = 0)$ ground level. Similarly, irradiation with σ^- polarized light leads to the population of the single magnetic substate with $m_F = -1$. Excitation into the $2p\ ^2P_{3/2}$ fine-structure level always populates more than one magnetic substate, since the hyperfine structure is not resolvable ($\Delta\nu_{\rm hfs} \sim 24$ MHz, whereas the natural width of 2^2P is $\Delta\nu \sim 94$ MHz). Also, using the $1s\ ^2S_{1/2}, F = 1$ as the initial level leads always to the population of more than one magnetic substate in the 2^2P intermediate level.

After the atom is electronically polarized in this way this polarization is transferred to the nucleus via the hyperfine interaction. Subsequent photoionization of these atoms by a second circularly polarized photon results in the production of nuclear-spin-polarized protons with 100% degree of polarization, polarized parallel or antiparallel to the propagation direction of the light beams, depending on the helicity of the light polarization. A detailed calculation of this two-step photoionization process can be performed using second-order perturbation theory. The probability $W_{J'_F}$ that the proton nuclear-spin component $|m_I\rangle$ is parallel to the direction of the light beams is then given by (assuming right-circularly polarized light)

$$W_{J'_F}(m_I) = g \sum_{(m_{F'},l)} \sum_{(J,m_J)} \left| \sum_{(F',m_{F'})} \langle ElJm_J | \langle \tfrac{1}{2} m_I | P_1 | 2^2P_{J'}, F'm_{F'} \rangle \langle 2^2P_{J'}, F'm_{F'} | P_1 | 1^2S_{1/2}, Fm_F \rangle \right|^2 ,$$

where $P_1 = 2^{-1/2}(P_x + iP_y)$ is the spherical component of the momentum operator $\mathbf{P}$, and g is a proportionality factor independent of J', F, and m_F. $\langle ElJm_J|$ characterizes the emanating photoelectron. The degree of nuclear polarization of the proton is then given in the usual way by

$$P_{J'_F} = \frac{W_{J'_F}(\uparrow) - W_{J'_F}(\downarrow)}{W_{J'_F}(\uparrow) + W_{J'_F}(\downarrow)} .$$

This quantity depends on the total angular momentum F of

the initial $^2S_{1/2}(F)$ state and on the electronic angular momentum J' of the intermediate $^2P_{J'}$ state. As a result we obtain for right-circularly polarized light, in agreement with other recently published approaches,[12,13]

$$P_{J'=1/2,F=0} = +1 \ ,$$

$$P_{J'=3/2,F=0} = -\frac{7}{11} \ ,$$

and

$$P_{J',F=1} = -\frac{1}{3} P_{J',F=0} \ .$$

The "+" sign denotes polarization parallel, the "−" sign antiparallel to the wave vector of the light beams. It is apparent that the two-photon ionization process with $^2S_{1/2}(F=0)$ as the initial and $^2P_{1/2}(F=1)$ as the intermediate level produces 100% polarized protons. In the case of hydrogen the photoelectron is emerging as a d wave. Contributions to the ionization from s-wave photoelectrons are found to be zero, in agreement with Ref. 13. Such s-wave photoelectrons, which could cause interferences in the photoionization and thereby lead to a diminution of the proton polarization, are possible for linearly polarized ionization light and also for atoms other than hydrogen. Changing the helicity of the light beams to left-circular polarization reverses the signs in the calculation and also in the results, thus reversing the polarization direction of the protons. This is conveniently accomplished without changing the excitation frequency of the laser.

The experimental apparatus was similar to that described by us previously.[14] For our purposes, tunable vuv radiation for excitation of the Lyman-α transition at $\lambda = 121.567$ nm was produced by third harmonic generation in krypton gas, as described in detail previously.[14,15] Tunable light near $\lambda = 364.7$ nm, i.e., the fundamental of the Lyman-α, was obtained by frequency mixing the nearly Fourier-transform-limited output of the tunable dye laser ($\Delta\nu \sim 100$ MHz) (Ref. 16) operating around $\lambda = 554.6$ nm with that of a Nd:YAG laser ($\lambda = 1064$ nm) in a deuterated potassium dihydrogen phosphate (KD^*P) crystal. The second harmonic of this Nd:YAG laser served to pump the dye laser. The bandwidth of the Nd:YAG laser was also narrowed to the Fourier-transform limit ($\Delta\nu \sim 65$ MHz) by inserting a 3.5-mm air-spaced etalon (finesse ~ 15) and a 13-mm solid fused-quartz etalon (finesse ~ 5) into the cavity. We also extended the mode buildup time by electronically controlling the opening on the Q switch. A similar system has been described in detail recently.[17] Due to its unstabilized resonator, the output frequency of the Nd:YAG laser varied occasionally between two longitudinal cavity modes with a frequency separation of about 250 MHz. After mixing this Nd:YAG laser radiation with the dye-laser radiation about 3-mJ linearly polarized light at $\lambda \sim 365$ nm in a 5-ns-long pulse was obtained. The pulse repetition rate was 10 Hz.

This radiation was focused with a 70-mm focal length lens into a cell containing Kr at a pressure of 130 mbar. About 3×10^9 linearly polarized vuv photons were generated, corresponding to a pulse energy of about 5 nJ. The spectral bandwidth of this tunable vuv radiation was about 700 MHz. The vacuum ultraviolet light and the generating fundamental at $\lambda = 364.7$ nm, which served to ionize the excited 2^2P atoms, were collimated by a MgF_2 lens and directed into the ionization region of a quadrupole mass filter (length ~ 8 mm). The diameter of the light beams in the interaction region were 1.3 and 5 mm for the exciting and ionizing radiation, respectively. A MgF_2 quarter wave plate for the production of circularly polarized light at Lyman-α was placed between the MgF_2 lens and the interaction region. The plate had a measured transmission of $(37 \pm 5)\%$ at Lyman-α. Hydrogen atoms were produced by a cooled microwave discharge in pure H_2. The gas was expanded through a capillary of 10-mm length and 1-mm diameter into the first stage of a differentially pumped vacuum system. The second stage contained the interaction region with the mass filter. A skimmer with a 0.25-mm-diameter opening placed 49 mm away from the capillary orifice defined a collimated atomic hydrogen beam with a residual Doppler width in the interaction region of $\Delta\nu_D \sim 550$ MHz at Lyman-α. In this setup the H atom density in the beam at the interaction region was about 6×10^9 cm^{-3}. The mass spectrometer was tuned to transmit mass $m/e = 1$ amu, and the signal from a secondary electron multiplier (SEM) was measured by a gated integrator, before being displayed on an x-y recorder.

Figure 1 shows the H^+ ion signal recorded as the exciting laser frequency is tuned across the Lyman-α transition. As can be seen, both fine-structure lines $1s\,^2S_{1/2} \rightarrow 2p\,^2P_{1/2}$ and $1s\,^2S_{1/2} \rightarrow 2p\,^2P_{3/2}$ are resolved further into their hyperfine components. This splitting reflects the hyperfine structure of the $1s\,^2S_{1/2}$ ground state with a separation of about 1420 MHz. Thus, the resonance observed at the relative position of 11 GHz (denoted by the arrow) reflects the excitation and subsequent ionization of the ($2p\,^2P_{1/2}$, $F=1$, $m_F=+1$) state, producing protons with nuclear-spin polarization parallel to the direction of the light beams. The degree of polarization, however, could not be verified in this simple apparatus. The theoretically expected[18] line positions are indicated in the lower part of Fig. 1. The measured line intensities of the hyperfine components correspond to the theoretically calculated relative line strengths in the ($1^2S_{1/2} \rightarrow 2^2P$) Lyman-$\alpha$ transition.[19] We thus conclude

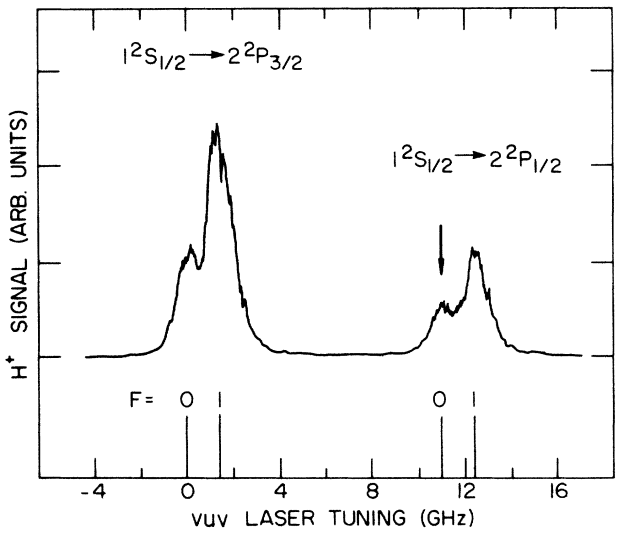

FIG. 1. High-resolution scan of the exciting right-circularly polarized vuv laser across the hydrogen Lyman-α line. Laser bandwidth was about 700 MHz, scan time ~ 20 min, integration time constant 3 s. In the lower part the expected positions of the transitions are indicated. The arrow denotes the transition where 100% spin polarized protons can be produced.

that within the experimental uncertainties the ionization probabilities of the magnetic substates in 2^2P populated within each line are about equal, in agreement with theoretical predictions.[20,21]

The overall two-step ionization probability of a hydrogen atom can be calculated by a rate equation model, which takes losses in the intermediate state due to spontaneous and stimulated emission into account,[22] which is necessary due to the high spontaneous decay rate of the 2^2P state. Under the given experimental conditions with $I_1 = 2.5 \times 10^{11}$ vuv photons per cm^2 and $I_2 = 2.5 \times 10^{16}$ ionizing photons per cm^2 at the interaction region, an ionization probability for ground-state hydrogen atoms of about 1.6% is calculated. This value already compares favorably with the most sophisticated electron ionizers currently in use on polarized proton sources.[5] In this calculation we used an effective excitation cross section $\sigma_1 = 8 \times 10^{-13}$ cm^2 and an ionization cross section of the 2^2P state of $\sigma_2 = 1.35 \times 10^{-17}$ cm^2. At the given H-atom density of 6×10^9 cm^{-3} and an effective ionization volume of 1.3×10^{-3} cm^3 a production of 1.2×10^5 nuclear-spin-polarized protons is estimated. From the measured signal and the previously determined transmission of the ion optics and the quadrupole,[15] we deduce a production of 7.5×10^4 polarized protons, in good agreement with the expected value. These protons are produced in about 5 ns, yielding a peak current of about 2.4 μA.

When compared with established sources, this current seems to be rather low—the ground-state sources deliver about 100 μA and the metastable state sources about 1 μA time-averaged proton current[5,6]—however, these results thus far are preliminary. The ion intensity can be improved by designing an appropriate reaction chamber, improving the Lyman-α generation, and using a second, more powerful laser for the ionization step. The necessary techniques are readily available. A Lyman-α light intensity of 40 nJ, which has already been obtained in our laboratory, together with moderate 30 mJ per pulse from the third harmonic of a second Nd:YAG laser will increase the overall ionization probability to about 73%. With such improvements the polarized proton production can potentially be increased by at least three orders of magnitude. The clean ionization mechanism—only H atoms were ionized; no other ions, e.g., H_2^+ or H_3^+, were observed in the mass spectrometer—the high degree of polarization, the inherent short proton pulse, and the simple method of changing the polarization are promising features of this new polarized proton source. Finally, it should be noted that with the current spectral resolution polarized tritons can also be generated. For polarized deuterons the resolution has to be increased.

The authors are grateful to Professor K. H. Welge for his interest in this research, and to Professor V. W. Hughes, Professor H. Klar, and Professor H. Kleinpoppen for stimulating discussions.

[1] *International Symposium on High-Energy Physics with Polarized Beams and Polarized Targets,* edited by C. Joseph and J. Spoffer, Experientia Supplemento Series, No. 38 (Birkhäuser Verlag, Basel, 1981).

[2] *Polarization Phenomena in Nuclear Physics—1980,* AIP Conf. Proc. No. 69, edited by G. G. Olson, R. E. Brown, N. Jarmie, W. W. McNaughton, and G. M. Hale (AIP, New York, 1981).

[3] *Polarized Proton Ion Sources* (Ann Arbor, 1981), AIP Conf. Proc. No. 80, edited by A. D. Krisch and A. T. M. Lin (American Institute of Physics, New York, 1982).

[4] G. Clausnitzer, R. Fleischmann, and H. Schopper, Z. Phys. **144**, 336 (1956).

[5] For a review see, e.g., W. Grüebler and P. A. Schmelzbach, in Ref. 2, p. 848ff.

[6] For an overview see, e.g., V. W. Hughes, in Ref. 3.

[7] G. J. Witteveen, Nucl. Instrum. Methods **158**, 57 (1979).

[8] L. W. Anderson, Nucl. Instrum. Methods **167**, 363 (1979).

[9] E. A. Hinds, W. D. Cornelius, and R. L. York, Nucl. Instrum. Methods **189**, 599 (1981).

[10] For example, J. L. McKibben, in Ref. 2, p. 830ff; E. G. Colton, I. P. Auer, A. Beretvas, D. Hill, K. Nield, B. Sandler, H. Spinka, D. Underwood, Y. Watanabe, and A. Yokosawa, Nucl. Instrum. Methods **151**, 85 (1978).

[11] S. A. Wender, C. E. Floyd, T. B. Clegg, and W. R. Wylie, Nucl. Instrum. Methods **174**, 341 (1980).

[12] M. F. Fedorov, Opt. Commun. **26**, 183 (1978); N. B. Delone, B. A. Zon, and M. F. Fedorov, Zh. Eksp. Teor. Phys. **76**, 505 (1979) [Sov. Phys. JETP **49**, 255 (1979)].

[13] R. Parzynski, Opt. Commun. **34**, 361 (1980).

[14] H. Zacharias, H. Rottke, J. Danon, and K. H. Welge, Opt. Commun. **37**, 15 (1981).

[15] H. Zacharias, H. Rottke, and K. H. Welge, Opt. Commun. **35**, 185 (1980).

[16] R. Wallenstein and H. Zacharias, Opt. Commun. **32**, 429 (1980).

[17] Y. K. Park and R. L. Byer, Opt. Commun. **37**, 411 (1981).

[18] G. W. Erickson, J. Phys. Chem. Ref. Data **6**, 831 (1977).

[19] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge Univ. Press, Cambridge, 1957).

[20] A. Burgess and M. J. Seaton, Mon. Not. R. Astron. Soc. **120**, 121 (1960).

[21] G. Peach, Mem. R. Astron. Soc. **71**, 13 (1967).

[22] H. Zacharias, R. Schmiedl, and K. H. Welge, Appl. Phys. **21**, 127 (1980); D. S. Zakheim and P. M. Johnson, Chem. Phys. **46**, 263 (1980); H. Rottke and H. Zacharias, J. Chem. Phys. (to be published).