A STUDY OF SPIN-EXCHANGE POLARIZATION TRANSFER IN HYDROGEN-RUBIDIUM COLLISIONS

A.N. Zelenski and S.A. Kokhanovski

Institute for Nuclear Research, Russian Academy of Sciences, 117312 Moscow

and

TRIUMF, 4004 Wesbrook Mall, Vancouver, B.C. V6T 2A3

ABSTRACT

The results of experimental studies of basic limitations on optical pumping of high density rubidium vapor are presented. A maximum 40% proton polarization was obtained in a spin exchange optically pumped polarized H^- ion source using a 30 cm long dry-film coated cell. About 50% polarization was measured with a 90 cm long copper uncoated cell at a Rb thickness of 8×10^{14} atoms/cm². A pulsed Ti:sapphire laser was used for optical pumping of Rb with up to 1 kW pulsed power, 200 μ sec pulse duration and 10-15 GHz linewidth.

INTRODUCTION

The recent progress made in the development of optically pumped polarized H^- ion sources is very impressive. The polarization in excess of 80%, which was obtained from the dc TRIUMF source, is actually higher than was expected taking into consideration all possible depolarization processes. The record current of polarized H^- - over 20 μ A in dc mode (TRIUMF)¹ and above 400 μ A in pulsed operation (INR, Moscow)² - was also obtained from optically pumped sources. Furthermore, the full capabilities of optically pumped sources have not been realized as yet, due to the very low efficiency with which the laser power is used. A 5 W Ti:sapphire laser used for optical pumping of alkali-metal vapor yields an electron polarization production rate of up to 10^{19} atoms/sec and a theoretical limit on equivalent polarized proton current of up to 1 A. This may be a way of producing multiampere polarized beams for fusion applications. The much smaller operational currents mentioned above are due to short polarization relaxation times and limits on production of low divergence high current proton beams. It is clear that the INR-type OPPIS in pulsed, low duty factor operation is capable of producing at least 1-2 mA of polarized H^- within a normalized emittance of 1.5π mm-mrad, suitable for high energy accelerators.³ The possibility of further improvement will be limited by the space-charge effect of Rb ions on the proton beam, which is apparently already significant in the dc OPPIS.

A possible way to avoid the above limitation and better realize the capabilities of optical pumping is to use the spin-exchange technique of polarization.⁴ Polarization transfer through spin-exchange collisions at thermal energies is well known. The thermal cross sections for alkali-alkali spin-exchange are above 10^{-14} cm². Recently, hydrogen and deuterium atoms were successfully polarized in collisions with optically pumped potassium. An important calculation was done

by Swenson *et al.*,⁵ it showed that the spin-exchange cross section of hydrogen-alkali collisions is $(1 - 3 \times 10^{-15} \text{ cm}^2)$ at hydrogen beam energies of 1-10 keV. The conclusion was that spin-exchange should slightly increase the polarization in OPPIS and is also important for the "collisional pumping" technique of polarization. "Collisional pumping" of proton polarization due to the hyperfine electron-proton interaction can occur only in a low magnetic field.⁶ The maximum density of optically-pumped alkali vapor is strongly limited by radiation trapping, as was shown by Tupa *et al.*,⁷ and to polarize a 10^{15} atoms/cm² thickness we need a 10 m long cell at low field. But if we use spin-exchange only for electron polarization in a high magnetic field and then use the conventional Sona method of polarization transfer, the technique looks feasible, since a 5×10^{12} atoms/cm³ polarized density has already been achieved.

A layout of the spin-exchange optically-pumped polarized ion source is presented in Fig. 1. In the original INR source, a neutral hydrogen beam was injected from an external source into a solenoid, where a helium ionizer cell and an optically pumped cell are situated.⁸ The only difference between the INR charge-exchange source and spin-exchange source is the absence of the helium ionizer cell in the latter. The hydrogen beam is neutral and we avoid any possibility of emittance degradation caused by Rb ion space-charge. Spin-exchange occurs between the ground states of the hydrogen and alkali atoms, and therefore only a 5-8 kG magnetic field is required for optical pumping of high density vapour, in contrast with the charge-exchange OPPIS, where over 20 kG is required to prevent depolarization due to the spin-orbital interaction in excited states.

The results of calculations of H-Rb and H-Cs spin-exchange polarizations at a hydrogen beam energy of 1-2 keV are presented in Fig. 2. The calculations show that production of 90% electron polarization of hydrogen atoms (and consequently about 85% proton polarization) requires at least a 10^{15} atoms/cm² thickness of optically pumped Rb vapour with polarization in excess of 95%. The ability to optically pump high density alkali vapour is crucial for the feasibility of spin-exchange sources. In contrast, the charge-exchange source can produce high polarization even at low Rb thickness, if background is kept low by reducing neutralization on residual gas, or by energy separation. For example, at the TRIUMF OPPIS, polarization drops only at a Rb cell thickness less than 2×10^{13} atoms/cm². The experimental limit on optical pumping of high density vapour is about 5×10^{12} atoms/cm³ and on thickness about 10^{14} atoms/cm² (with a 20 cm long sodium cell in pulsed mode of operation at INR, Moscow⁸ and KEK⁹). Increasing the cell length is a simple way to get more thickness at limited density (supposing laser power is unlimited, as in the case of pulsed operation), but in practice it will limit the polarized current, because losses of the hydrogen beam in a 200 cm cell will be too high. (The divergence of the hydrogen beam from the best sources is about 20 mrad at 1-2 keV beam energy). So, to realize the advantages of spin-exchange polarization it is necessary to increase the density of optically pumped vapor to at least $(1 - 2) \times 10^{13}$ atoms/cm³. The first experimental results on spin-exchange polarization in hydrogen-sodium collisions were presented at the last workshop at KEK.⁴ Recent progress has been made due to the switch to optical pumping of Rb and pulsed Ti:sapphire lasers.

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Fig. 1. Schematic layout of the spin-exchange polarized ion source: 1) source of primary protons; 2) focusing solenoid; 3) neutralizing cell (H_2 -Xenon); 4) optically pumped Rb cell; 5, 8 pulsed solenoids; 6) deflection plates; 7) sodium ionizer cell for H^- ion production (or He gas cell for proton production); 9) bending magnets; 10) $\lambda=795$ nm pulsed Ti-Sp laser; 11) polarimeter sodium cell; 12) spin filter; 13) detector for Lyman-Alpha photons; $CF_{1,2,3}$ - Faraday cylinders.

Fig. 2. Calculated spin exchange cross sections as a function of the energy for fast H^0 atoms incident on H, Na, Rb and Cs. (from Ref.⁵)

EXPERIMENTAL SETUP FOR SPIN-EXCHANGE POLARIZATION STUDIES

By using a dry-film wall coating the polarization relaxation time was increased to up to 600 μ sec. From the calculation of radiation trapping it was expected that the maximum vapor density in the optically pumped cell, given a 600 μ sec relaxation time, would be at least 5×10^{13} atoms/ cm^3 (see Ref.¹⁰). The experimental results are apparently in contradiction with this calculation, and probably there is another polarization loss process, which will be discussed later.

The experiments were done at the test-bench of the INR pulsed OPPIS. Downstream of the optically pumped spin-exchange cell everything has the same

configuration as in the charge-exchange source. Electron polarization is transferred to the proton by a Sona transition and then H^- ions are produced by second electron capture in the alkali-metal vapor ionizer cell. It seems that the optimal energy of the primary neutral hydrogen beam for spin-exchange sources will be about 1-2 keV. At this energy the spin-exchange cross sections for H-Rb and H-Cs collisions are up to 3×10^{-15} cm^2 . Also at this energy very intense neutral hydrogen sources are available, which were developed for plasma diagnostics in fusion research.¹¹

In experiments on spin-exchange polarization we used a neutral hydrogen beam injector (the prototype was developed at Budker INP, Novosibirsk),¹¹ which yielded at 7 keV up to 30 mA of equivalent neutral current through the optically pumped cell and helium ionizer. The source of primary protons was optimized for an extraction voltage of 7-10 keV and produced substantially lower current at 2-2.5 keV beam energy, where we did most of the experiments on spin-exchange polarization. We had about 200-400 μA of polarized proton beam, which was enough for polarization measurements even at low duty factor. The source operates at a 1 Hz repetition rate, limited by the pulsed solenoid cooling system and 100 μ sec pulse duration. For proton polarization measurements we used our conventional technique, based on analysis of metastable hydrogen atoms which are produced by neutralization of polarized protons in a polarimeter sodium cell.⁸

Most of the experiments on spin-exchange polarization were done with a 35 cm long and 1.4 cm diam Rb cell. The cell was made from stainless steel with a copper inner tube (liner). The copper liner was mechanically and electrochemically polished. We used a procedure of dry-film coating described in detail by Swenson *et al.*¹² Rubidium vapor from a 30 cm long reservoir enters the cell through a 0.1 cm wide, 25 cm long slit. In the first experiments Rb was loaded directly into the reservoir. It was very inconvenient and unsafe handling the long cell, loaded with very reactive Rb. When exposed to air Rb is coated by an oxide layer, and to break through this layer it is necessary to raise the cell temperature much higher than operational. The Rb density then increases suddenly, sometimes causing problems for further cell operation, in particular, often destroying the dry-film wall coating. We later invented "self-opening" Rb containers. Such a container consists of two pieces of stainless steel tube, each closed at one end by a copper plug. The two tubes fit closely together to reduce contact of Rb with air, and a compressed spring is installed between them. After loading with Rb in a glove box, the tubes are fastened together by two strips of copper foil soldered on with low melting point alloy. Once the containers are loaded, we can handle them easily, since they are almost hermetically sealed. Inside the reservoir, the solder melts at 70-90°C and the spring forces the tubes apart, well below the operational temperature of 150°C. For a 35 cm cell we used 4 containers to provide a homogeneous distribution of Rb density along the cell. This technique was even more important for experiments with a 90 cm cell.

We used a Faraday rotation technique for measuring the Rb vapor thickness. A small movable mirror, installed on the source axis between the Rb cell

and the proton neutralizer cell, directed a laser beam to an analyzing polarizer after the beam had passed through the Rb cell. We used the pumping laser as a probe for thickness measurements, and therefore could not use the Faraday rotation technique for polarization measurements. The rubidium and hence proton polarization were varied from pulse to pulse by scanning the laser wavelength with a tilting intracavity etalon. The laser wavelength was measured every pulse by a wavemeter, based on analysis of the interference pattern from a Fizeau interferometer. The laser wavelength was tuned by measuring the laser power absorption curve.

EXPERIMENTAL RESULTS OF THE POLARIZATION STUDIES

We studied the polarization relaxation time by measuring the polarization as a function of delay time between laser pulse and measuring gate. The relaxation time strongly depends on the cell wall surface conditions. The 600 μsec relaxation time has been measured with a fresh dry-film coated cell and 200 μsec for a fresh uncoated copper liner (see Fig. 3).

It follows from calculations of radiation trapping in optically pumped high density vapor, that even at infinite laser power there is a maximum polarization limit which depends on the polarization relaxation time.¹⁰ For a 600 μsec relaxation time, radiation trapping limits polarization above a Rb density of about $(3-5) \times 10^{13}$ atoms/cm³. The results of proton polarization measurements as a function of Rb thickness are presented in Fig. 4. In contrast with calculations, polarization drops at a thickness above 5×10^{14} atoms/cm², corresponding to a density of 1.5×10^{13} atoms/cm³. Apparently, there are other depolarization processes besides radiation trapping. For example, at high density and high pumping power, Rb atoms in excited states become important. We discovered very efficient ionization in collisions between sodium atoms in excited states.¹³ We can roughly estimate the density of excited states from measurements of

Fig. 3. Rb polarization relaxation time for copper liner with fresh dry-film coating. Measuring gate is 20 μsec .

Fig. 4. Proton spin-exchange polarization dependence on optically pumped Rb vapour thicknesses. Short 35 cm cell with dry-film coating. Long 90 cm cell uncoated. Magnetic field - 8 kG.

absorbed laser power and the time of Rb 5p-state spontaneous decay. The result is an excited state density of 10^{12} atoms/cm³ at a Rb density of 10^{13} atoms/cm³. The process of excitation exchange might be the major depolarizing process at this density, since the cross section of the excitation exchange process:

$$Rb(5P_{1/2}) + Rb_j \rightarrow Rb(5P_{3/2})$$

is about $10^{-11} - 10^{-12}$ cm² (see Ref.¹⁴). After exchange, the polarized atom can lose polarization by spontaneous decay. An increase in relaxation time reduces both the pumping power and the rate of excited states production. But excitation exchange limits the optically pumped cell density. Apparently, the density limit of $(1-2) \times 10^{13}$ atoms/cm³ is lower than we expected from radiation trapping calculations alone.

To complete our study of optical pumping of monatomic vapor, we built a 90 cm long cell. The copper liner's inner diameter was 1.6 cm. The cell reservoir was loaded with 8 "self-opening" Rb containers. The cell temperature was controlled by circulation of glycerin at up to 180° C. The results of polarization measurements with the long cell are presented in Fig. 4. We obtained up to 50% proton polarization at a Rb vapor thickness of 6×10^{14} atoms/cm². This result is in good agreement with extrapolation of data from the 35 cm uncoated cell. Of course, the real advantage of the longer cell is only realized with a dry-film wall coating. Unfortunately, we have not yet the equipment for wall-coating the long cell.

OPTICAL PUMPING OF ALKALI-METAL VAPOR MIXTURES

As mentioned above, polarization transfer in spin-exchange collisions is a more efficient process at thermal energy, due to the higher cross section. Therefore when one species in a mixture of two alkali-metal vapors is optically pumped,

the other also becomes polarized due to spin-exchange collisions. Initial discussions of using a mixture of alkali-metal vapors in the optically pumped cell of a polarized ion source only took into account the buffering effects of the second species.¹⁵ It was then realized that optical pumping of mixtures reduces radiation trapping depolarization, since we can keep the density of optically pumped vapor below 10^{13} atoms/cm³ and nevertheless get high total density (5×10^{13} atoms/cm³) due to a high buffer vapor density. The use of a mixture also avoids depolarization due to excitation exchange, since the cross section of excitation exchange between different alkalis should be a few orders less than the resonant cross section between identical atoms. In our first experiments on optical pumping of potassium-sodium mixtures, we obtained 7×10^{14} atoms/cm² total mixture thickness with 70% polarization.¹⁶ Apparently the best choice of alkalis is the rubidium-cesium mixture. Optical pumping of Rb vapor by Ti:sapphire lasers is very efficient and well studied. The spin-exchange cross section of Rb-Cs collisions is very high (2×10^{-14} cm²). Also, the spin-exchange cross sections are highest for H-Rb and H-Cs collisions at 1-2 keV hydrogen beam energy. The use of a Rb-Cs mixture allows low temperature operation of the optically pumped cell and the use of a dry-film wall coating for increasing the polarization relaxation time.

The cell for studying Rb-Cs mixture optical pumping is shown in Fig. 5. Rb and Cs are loaded in two reservoirs having independent temperature control, allowing separate control of the Rb and Cs fluxes and hence of the density ratio in the cell. The dry-film coated copper liner is 30-50° C hotter than the reservoirs to prevent any condensation on the wall coating. We are planning to study optical pumping of Rb-Cs mixtures in collaboration with TRIUMF.

Fig. 5. Cell for experiments on optical pumping of Rb-Cs mixture. 1) stainless steel cell body; 2) copper liner with dry-film coating; 3) "self-opening" container; H) heaters; C) cooling tubes.

ESTIMATION OF CURRENT FROM SPIN-EXCHANGE SOURCES

The hydrogen beam undergoing polarization is neutral in spin-exchange sources and therefore current estimation is quite simple and directly follows from experimental results on neutral injector development and the source geometry. We will make all estimates for production of polarized H⁻ ions, since that is

the best choice for high energy accelerators, but as is well known, the OPPIS is capable of producing ten times higher polarized proton beam by using ionization in a helium cell.

The geometry of the ionizer is determined by the specified polarized beam emittance, since the divergence of the final polarized beam is produced mostly when H⁻, created by charge exchange inside the ionizer magnetic field, leaves the solenoidal field.¹⁷ The initial neutral beam emittance is negligible. At typical ionizer parameters - $B=1.5$ kG ionizer solenoid magnetic field and $2R=1.5$ cm diam of ionizer cell - the emittance increase is equal to 1.3π mm-mrad, close to the maximum emittance acceptable for high energy accelerators. The typical length of the ionizer cell is about 30 cm, which could be reduced to 15-20 cm by using a jet ionizer. The distance between the ionizer and optically pumped cell varies from 25 cm (INR OPPIS with small 4.2 cm bore of pulsed solenoid) to 50 cm (LAMPF, TRIUMF OPPIS with large 15 cm bore of superconducting solenoid). The magnetic field of 5-8 kG required for the spin-exchange source might be produced by a conventional solenoid, allowing a more compact design than a superconducting solenoid. We assume a 30 cm distance between the optically pumped cell and the ionizer for the spin-exchange source. The length of the optically pumped cell should be in the range of 30-100 cm. A short cell would be used in the case of using alkali mixtures, the long cell with monotonic Rb vapor. The diameter of the optically pumped cell is limited by the available laser power in dc operation. A reasonable choice is 1.0 cm. In pulsed operation the laser power is not a limitation and especially for the case of a long cell and converging primary proton beam it is reasonable to use a larger (2-3 cm) diam cell. The absence of a strong limitation on the diameter of the optically-pumped cell is another important advantage of spin-exchange polarization, since in the OPPIS with ECR source, increasing the cell entrance diameter raises gas loading and neutralization on residual gas, consequently reducing the polarization.

The high intensity convergent atomic hydrogen beams are produced by neutralization of a high-brightness proton beam just after a focusing solenoidal lens.¹¹ (A convergent proton beam might also be produced by using a spherical extraction system). In such a way, up to 1 A/cm^2 of equivalent neutral hydrogen current was obtained at a distance of 100 cm from the source at 1 keV beam energy in pulsed operation at 10 Hz repetition rate and 200 μsec pulse duration.¹⁸ As discussed above, the full length of the optically pumped cell and ionizer assembly should be 110-160 cm. The beam focus should be at the exit of the optically pumped cell or closer to the Sona transition region. A small beam diameter at the Sona region ensures that no polarization is lost there. At least 100 mA of equivalent neutral current will pass through a 1.5 cm diam ionizer cell. Rb vapor appears to be the best choice for the ionizer cell. The H⁻ yield from a Rb ionizer should be about 20% and the resulting polarized H⁻ current will be 20 mA. That is close to unpolarized source currents, and development of such a source will provide great opportunities for high energy spin-physics experiments.

CONCLUSION

Optically pumped polarized H^- ion sources now produce higher current than the best atomic beam sources in both pulsed and dc operation, with the promise of much higher capabilities to come. We believe that the development of the spin-exchange polarization technique will lead to polarized H^- currents in excess of 10 mA. Detailed study of the processes involved in optical pumping of high density alkali-metal vapor and of hydrogen beam interaction with such media must be accomplished to fully realize the power of the optical pumping technique.

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