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OPTICAL PUMPING PRODUCTION OF SPIN POLARIZED HYDROGEN

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SEPTEMBER 1984

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UNDER CONTRACT DE-AC02-75-OR-00171

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PPPL--2125
DE85 002130

I. INTRODUCTION

There has been much interest recently in the production of large quantities of spin polarized hydrogen in various fields, including controlled fusion, quantum fluids, high energy, and nuclear physics. One promising method for the development of large quantities of spin polarized hydrogen is the utilization of optical pumping with a laser. Optical pumping is a process in which photon angular momentum is converted into electron and nuclear spin. The advent of tunable CW dye lasers (~ 1 watt) allows the production of greater than 10^{18} polarized atoms/sec. We have begun a program at Princeton to investigate the physics and technology of using optical pumping to produce large quantities of spin polarized hydrogen. Initial experiments have been done in small closed glass cells. Eventually, a flowing system, open target, or polarized ion source could be constructed.

An example illustrating optical pumping¹ is shown in Fig. 1 for ^{87}Rb which possess a nuclear spin of $I = 3/2$. The rubidium vapor is illuminated with circular polarized D1 resonance light (795 nm). Conservation of angular momentum requires that for excitation to the $P_{1/2}$ state the selection rule $\Delta m_F = +1$ applies. The excited atoms quickly decay back to

the ground state with the selection rule $\Delta m = \pm 1, 0$ weighted by the respective branching ratios. The net result is the increase in the absolute value of the angular momentum, or population of the $F = 2$, $M_F = \pm 2$ state (dependent on the use of left versus right-hand polarized light). Relaxations will repopulate the other sublevels. The steady-state electron spin polarization S_R will be given by

$$S_R = \frac{\Gamma_{\text{opt}}/2}{\Gamma_{\text{opt}} + \gamma_R} \quad (1)$$

where Γ_{opt} is the optical pumping rate, and γ_R is the rubidium relaxation rate. The optical pumping rate will be approximately

$$\Gamma_{\text{opt}} \sim I \sigma_{\text{ABS}} \quad (2)$$

where I is the flux of photons, and σ_{ABS} ($\sim 10^{-11} \text{ cm}^2$) is the absorption cross section. At 795 nm and 1 watt/cm^2 , $\Gamma_{\text{opt}} \sim 4 \times 10^7 \text{ sec}^{-1}$. Typical γ_R is 100 sec^{-1} and 1 sec^{-1} for uncoated and coated cells, respectively. Thus, the rubidium polarization can exceed 99%, and the polarization rates of $\sim 10^{18} \text{ sec}^{-1}$ should be feasible. Atomic hydrogen cannot be directly polarized in a similar manner because of the lack of intense Lyman alpha light sources, good polarizers, and filters. Fortunately, spin exchange collisions allow polarization of hydrogen² and other species. After the rubidium is polarized, it can spin exchange with hydrogen atoms

where the arrows signify the electron spins. Spin exchange cross sections between paramagnetic species are large, being of the order $\sigma_{SE} \sim 10^{-14} \text{ cm}^2$. After the collision, the hyperfine interaction will couple the electron and nuclear spins of the hydrogen atom, resulting in a transfer of electron spin to the nucleus. After several spin exchange collisions, the hydrogen atom will eventually become both electron and nuclear spin polarized. In steady state, the hydrogen spin will be approximately

$$S_H \sim \frac{[Rb] v \sigma_{SE}}{[Rb] v \sigma_{SE} + \gamma_H} S_R \quad (4)$$

where $[Rb]$ is the rubidium density ($\sim 10^{11} - 10^{13} \text{ cm}^{-3}$), v is the relative velocity ($\sim 10^5 \text{ cm sec}^{-1}$), and γ_H is the hydrogen relaxation rate. Equation (4) is an approximation where the recombination time of hydrogen atoms is assumed to be long. For a coated cell where $\gamma_H \sim 1 \text{ sec}^{-1}$ and at a density of $[Rb] = 10^{12} \text{ cm}^{-3}$, Eq. (4) yields $S_H = 0.999 S_R$. Thus for a cell being pumped with a laser, both the rubidium and hydrogen should be almost completely polarized.

2. EXPERIMENTAL RESULTS

Figure 2 shows a schematic diagram of the apparatus. A Coherent Ring Dye Laser (Model 699-21) is operated with LD700 dye and pumped by a Spectra Physics (Model 171-01) Krypton ion pump laser. About 600 mW of broad band ($\Delta f \sim 7 \text{ GHz}$) and 400 mW single frequency ($\Delta f \sim 1 \text{ MHz}$) power are available at the first rubidium resonance wavelength of 795 nm. The laser light is circularly polarized with a linear polarizer and quarter wave plate. In addition to the laser, a resonance lamp has been utilized for pumping. The

advantage of a resonance lamp is the simplicity of operation, and the fact that the lamp operates at the shot noise limit although the intensity is low (microwatts cm^{-2}). The Rb density can be controlled by changing the pyrex cell temperature with a typical temperature of 100°C yielding $[\text{Rb}] \sim 5 \times 10^{12} \text{ cm}^{-3}$. The circular polarized light optically pumps the rubidium vapor. As the vapor is polarized, the light transmission will increase with a decrease in the fluorescence. In addition to pumping the rubidium vapor, the laser (lamp) can be used to probe the polarization. The cell is located in a few gauss magnetic field which defines the axis of quantization and provides a small Zeeman splitting (see Fig. 3). A radiofrequency field (few MHz) can be applied at either the rubidium or hydrogen Zeeman frequency causing depolarization. This depolarization leads to a reabsorption of pumping light, and therefore an increase in the fluorescence and an increase in the opacity of the vapor. Depolarization of the hydrogen depolarizes the rubidium due to spin exchange collisions. Hydrogen atoms are formed by a pulsed (10^{-4} sec) radiofrequency discharge. The Zeeman RF field is modulated (~ 10 - 100 Hz) with two discharge pulses/cycle. The signal is measured with a lock-in amplifier as the difference in transmission (or fluorescence) with and without the depolarizing RF field.

Figure 4 shows a typical rubidium signal obtained using a resonance lamp at a 9 gauss magnetic field. The quadratic Zeeman effect separates the individual transitions within the F manifolds. This quadratic Zeeman splitting allows convenient direct determination of the spin temperature and polarization. If spin exchange collisions are sufficiently rapid, the population of the sublevel m can be described by a spin temperature β ,

$$\rho(m) = \frac{e^{\beta m}}{\sum e^{\beta m}}.$$

Analysis of Fig. 4 yields a spin temperature of $\beta = 0.2$ or a rubidium electron or nuclear spin of about 10%. This polarization is in agreement with other measurements of γ_R , Γ_{opt} , and utilization of Eq. (1).

Figure 5 shows the observed Zeeman signals for pumping with a 2 mW laser beam which was expanded to 0.5 cm diameter. In this case, the hydrogen spin was determined to be about 10%. Similar measurements for rubidium signal yielded a polarization of 30%. The major reason why these polarizations are not larger is due to diffusion out of the laser beam and subsequent depolarization at the walls. This problem will be corrected in future work by utilizing coated walls which will substantially decrease the relaxation rates.

3. ULTIMATE PRODUCTION RATES

In this section, we examine some of the limitations imposed on the ultimate densities and polarizations which can be achieved using optical pumping-spin exchange to produce polarized hydrogen.

The first requirement is for the spin exchange rate $R_{SE} = [Rb]v\sigma_{SE}$ to exceed the hydrogen relaxation rate γ_H in order to maximize S_H (Eq. 4). The spin exchange rate R_{SE} can be varied by changing the temperature. For typical operating temperatures (50-150°C), the rates are $10^2 - 10^5 \text{ sec}^{-1}$. The hydrogen will relax due to wall collisions and volume relaxation. The wall relaxation rate can be reduced to about 1 sec^{-1} by coating the walls with a suitable compound such as paraffin, Teflon, etc. The volume

relaxation will be due to spin flip collisions

$$H(\uparrow) + H(\uparrow) \rightarrow H(\uparrow) + H(\uparrow), \quad (6)$$

where one hydrogen spin can create a fluctuating magnetic field which causes the other atom to flip its spin. The interaction of two dipoles at a separation r is given by the interaction energy V ,

$$V = \frac{(3\hat{\mu}_1 \cdot \hat{r})(\hat{r} \cdot \hat{\mu}_2) - \hat{\mu}_1 \cdot \hat{\mu}_2}{r^3} \quad (7)$$

$$\sim \frac{\mu_B^2}{r^3},$$

where μ_B is the Bohr magneton. The cross section for depolarization is estimated to be

$$\sigma_{\text{DEPOL}} \sim \left(\frac{V\tau}{h}\right)^2 a^2, \quad (8)$$

$$\sim 10^{-21} \text{ cm}^2.$$

The depolarization rate will be

$$R_{\text{DEPOL}} = [H]v\sigma_{\text{DEPOL}}. \quad (9)$$

There will be a similar depolarization rate due to spin orbit interactions. The requirement that

$$R_{EX} > R_{DEPOL} \quad (10)$$

leads to a requirement on the ratio of hydrogen and rubidium densities,

$$\frac{[H]}{[Rb]} < \frac{\sigma_{SE}}{\sigma_{DEPOL}} \sim 10^7. \quad (11)$$

Since Rb vapors of densities of 10^{12} cm^{-3} can easily be polarized, the hydrogen density should be less than about 10^{17} cm^{-3} for 99% polarization.

Another requirement to achieve high polarization is that the laser power must exceed the depolarization rates. The loss of spin L_R due to rubidium wall collisions will be

$$L_R = [Rb] V \gamma_R S_R, \quad (12)$$

and the loss of spin due to the hydrogen L_H will be

$$L_H = [H]^2 V v \sigma_{DEPOL} S_H + [H] V \gamma_H. \quad (13)$$

Since $\gamma_R, \gamma_H \sim 1 \text{ sec}^{-1}$, the dominant spin loss mechanism will be due to spin flips in the vapor which leads to the requirement on the laser photon intensity I ,

$$I > [H]^2 V v \sigma_{DEPOL}. \quad (14)$$

For a 1 watt/cm² laser and 50 cm³ volume, $[H] < 2 \times 10^{16} \text{ cm}^{-3}$. Thus, there appears to be an upper limit of about 10^{16} cm^{-3} on the hydrogen density that can be highly polarized.

4. CONCLUSIONS

Laser Optical Pumping-Spin Exchange appears to be an efficient technique for the production of large quantities of spin-polarized hydrogen. Experiments have been initiated using a laser and lamp to investigate this technique. Hydrogen and rubidium polarizations of 10 and 30 percent have been achieved. Future experiments will use coated cells which will reduce the relaxation rates and increase S_H and S_R . It is expected that ultimate hydrogen polarizations exceeding 99% should be obtainable at densities of 10^{16} cm^{-3} .

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy Contract No. DE-AC02-76-CHO-3073, Duke Power Company, Pennsylvania Power and Light, PSE&G, and Southern California Edison Company.

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FIGURE CAPTIONS

- FIG. 1. Optical Pumping of ^{87}Rb . Illumination of the vapor with right hand circular polarized light eventually populates the $F = 2, m_F = 2$ state.
- FIG. 2. Schematic diagram of the apparatus.
- FIG. 3. Zeeman splitting of the $F=1$ hydrogen ground states. The quadratic dependence on the magnetic field allows determination of the spin temperature β and hydrogen polarization. The rubidium Zeeman effect is similar except there are more levels involved.
- FIG. 4. Observed rubidium Zeeman transitions obtained with resonance lamp pumping. Ratio of adjacent transitions yields a polarization of about 10%.
- FIG. 5. Observed hydrogen Zeeman transitions obtained with laser pumping. Ratio of the two transitions yields a nuclear polarization of about 10%.

#83P0064

FIG. 1

84P0182

FIG. 2

#83P0065

FIG. 3

#84P0126

FIG. 4

FIG. 5

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