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Section A

The effect of radiation trapping on a high field spin exchange optically pumped target

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The effect of radiation trapping on an optically pumped spin exchange polarized atomic hydrogen or deuterium target operated in a high magnetic field is analyzed. It is shown that, if possible, it is desirable even in a high magnetic field to use an alkali density low enough that significant radiation trapping does not occur since fewer photons are needed on the average to polarize an atom. This permits one to obtain the maximum number of polarized hydrogen atoms per photon. Operation at a low alkali density requires both a high atomic hydrogen density and a low hydrogen polarization loss rate. It is also suggested that in a high magnetic field there may be advantages in using linearly polarized light for the optical pumping.

1. Introduction

The prospects of producing a polarized atomic hydrogen or deuterium beam by spin exchange optical pumping in a high magnetic field appear very promising. In recent high magnetic field spin exchange optical pumping experiments, Coulter et al. have obtained a polarized beam of atomic deuterium with a flux of 2.1×10^{17} atoms/s and with an atomic polarization of $73 \pm 3\%$ [1]. In a high magnetic field the average number of photons needed to polarize an alkali atom increases as the alkali density increases due to radiation trapping. This paper discusses the effect of radiation trapping on the spin exchange optical pumping of hydrogen or deuterium in a high magnetic field and discusses the problems to address in order to increase the polarized hydrogen or deuterium flux.

A beam of polarized atomic hydrogen or deuterium can be produced using spin exchange optical pumping as follows. We consider the polarization of hydrogen in our discussion but the concepts apply to deuterium as well. Both an alkali vapor and atomic hydrogen are flowing through a cell. The alkali atoms are polarized by optical pumping in a high magnetic field. The atomic hydrogen is polarized by alkali–hydrogen (A–H) spin exchange collisions. There are also hydrogen–hydrogen (H–H) and alkali–alkali (A–A) spin exchange collisions. The H–H and A–A spin exchange collisions leave the electron spin polarization of both the hydrogen atoms and alkali atoms unchanged. The electron spin polarization of the atomic hydrogen in the cell is lost either by relaxation collisions with other atoms and molecules, or with the cell walls, by recombination, or by flow of hydrogen atoms out of the cell. The alkali atoms do not lose a large fraction of their polarization by relaxation or by the flow out of the cell. The alkali atoms lose most of their polarization by transferring it to the hydrogen atoms via spin exchange collisions. Since radiation trapping affects the rate of optical pumping of the alkali atoms it thereby affects the obtainable rate of polarization of the hydrogen atoms. This paper discusses the effects of radiation trapping on the conditions that will maximize the flow of polarized hydrogen out of a spin exchange optically pumpted cell.

2. The rate equations for high field spin exchange optical pumping

We discuss optical pumping using σ^+ light with a wavelength corresponding to absorption from the $^2S_{1/2}$ ground level of the alkali to the lowest $^2P_{1/2}$ excited level of the alkali. The optical pumping works as follows. Absorption of σ^+ light excites alkali atoms out of the $^2S_{1/2}$, $m = -1/2$ state into the $^2P_{1/2}$, $m = 1/2$ state which decays spontaneously to the $^2S_{1/2}$, $m = -1/2$ state two-thirds of the time and to the $^2S_{1/2}$, $m = 1/2$ state one-third of the time. The net result is to depopulate the $^2S_{1/2}$, $m = -1/2$ state and increase the population of the $^2S_{1/2}$, $m = 1/2$ state thereby polarizing the alkali vapor. The excess population in the $^2S_{1/2}$, $m = 1/2$ state of the alkali atoms tends toward equalization either by relaxation collisions or by flowing out of the cell and being replaced by unpolarized atoms. However, these are not the primary mechanisms for the loss of polarization from the alkali vapor. Polarization leaves the alkali atoms primarily by spin exchange collisions with the hydrogen atoms. The polarization of the hydrogen atoms is lost by relaxation collisions, by recombination or by flow out of the cell. If we ignore the density of alkali atoms in the excited $^2P_{1/2}$ level then we need to consider four rate equations, one each for the density of alkali atoms in the ground level with $m = -1/2$ and $m = 1/2$ denoted by $n_{A\beta}$ and $n_{A\alpha}$, respectively, and one each for the density of hydrogen atoms in the ground level with $m = -1/2$ and $m = 1/2$ and denoted by $n_{H\beta}$ and $n_{H\beta}$, respectively. The four rate equations are the following:

$$\frac{dn_{A\alpha}}{dt} = \frac{R}{N} n_{A\beta} - n_{A\alpha} n_{H\beta} \langle \sigma v \rangle + n_{A\beta} n_{H\alpha} \langle \sigma v \rangle - \frac{n_{A\alpha} - n_{A\beta}}{2T_A}, \quad (1)$$

$$\frac{dn_{A\beta}}{dt} = \frac{R}{N} n_{A\beta} + n_{A\alpha} n_{H\beta} \langle \sigma v \rangle - n_{A\beta} n_{H\alpha} \langle \sigma v \rangle + \frac{n_{A\alpha} - n_{A\beta}}{2T_A}, \quad (2)$$

$$\frac{dn_{H\alpha}}{dt} = n_{A\alpha} n_{H\beta} \langle \sigma v \rangle - n_{A\beta} n_{H\alpha} \langle \sigma v \rangle - \frac{n_{H\alpha} - n_{H\beta}}{2T_H}, \quad (3)$$

$$\frac{dn_{H\beta}}{dt} = -n_{A\alpha} n_{H\beta} \langle \sigma v \rangle + n_{A\beta} n_{H\alpha} \langle \sigma v \rangle + \frac{n_{H\alpha} - n_{H\beta}}{2T_H}, \quad (4)$$

where R is the laser optical absorption rate in photons/s, N is the average number of photons needed to transfer an alkali atom from the $^2S_{1/2}$, $m = -1/2$ state to the $^2S_{1/2}$ $m = 1/2$ state, i.e. to polarize an alkali atom, $\langle \sigma v \rangle$ is the average over the velocity distribution of the product of the A–H spin exchange cross section times the A–H relative velocity, T_A is the alkali polarization loss time, and T_H is the hydrogen polarization loss time. The alkali and hydrogen polarization loss times are given by $1/T_A = 1/T_{AR} + 1/T_{AF}$ and $1/T_H = 1/T_{HR} + 1/T_{HF}$ where T_{AR} is the alkali relaxation time and T_{AF} is the average dwell time of an alkali atom in the cell, T_{HR} is the hydrogen relaxation time including the effect of recombination and T_{HF} is the average dwell time of hydrogen atom in the cell. The laser absorption rate is given by

$$R = \left(\frac{I_\nu}{h\nu} \bar{\sigma}_{ABS} \right), \quad (5)$$

where I_ν is the laser intensity in power per unit area, ν is the laser frequency and $\bar{\sigma}_{ABS}$ is the effective absorption cross section for the atomic line shape folded with the laser frequency distribution. Neither A–A nor the H–H spin exchange terms appear in the rate equation because these terms do not change the state populations.

The rate equations are simplified by rewriting them in terms of the alkali and hydrogen polarizations:

$$P_A = \frac{n_{A\alpha} - n_{A\beta}}{n_A} \quad (6)$$

and

$$P_H = \frac{n_{H\alpha} - n_{H\beta}}{n_H}, \quad (7)$$

where $n_A = n_{A\alpha} + n_{A\beta}$ and $n_H = n_{H\alpha} + n_{H\beta}$ are the total alkali and hydrogen densities, respectively. The spin up and spin down densities can be written as $n_{A\alpha} = n_A(1 + P_A)/2$, $n_{A\beta} = n_A(1 - P_A)/2$, $n_{H\alpha} = n_H(1 + P_H)/2$, and $n_{H\beta} = n_H(1 - P_H)/2$.

The rate equations rewritten in terms of the alkali and hydrogen polarizations are given by

$$\frac{dP_A}{dt} = \frac{R}{N}(1 - P_A) - \langle \sigma v \rangle n_H (P_A - P_H) - \frac{P_A}{T_A} \quad (8)$$

and

$$\frac{dP_H}{dt} = \langle \sigma v \rangle n_A (P_A - P_H) - \frac{P_H}{T_H}. \quad (9)$$

3. Analysis of spin exchange optical pumping in a high magnetic field

In the steady state eq. (9) leads to the result

$$\frac{P_H}{P_A} = \frac{n_A \langle \sigma v \rangle T_H}{1 + n_A \langle \sigma v \rangle T_H}$$

or

$$n_A \langle \sigma v \rangle T_H = \frac{P_H/P_A}{(1 - P_H/P_A)}. \quad (10)$$

Thus for a given alkali density n_A, if one requires a minimum value of P_H/P_A, T_H must have a minimum value that can be calculated if $\langle \sigma v \rangle$ is known. In order to make the discussion concrete consider the case where the alkali atom is sodium with an atomic mass of 23. In this case $\langle \sigma v \rangle = 2.3 \times 10^{-9}$ cm^3 s^{-1} at a temperature of 500 K [2]. The value of $\langle \sigma v \rangle$ depends only weakly on the temperature. Again in order to make the discussion concrete let us consider the case where $P_A = 7/8$ and $P_H = 3/4$ or greater. For these values of P_A and P_H it is seen that $P_H/P_A = 6/7$ or larger which requires that $T_H \geq 6/(n_A \langle \sigma v \rangle)$. Thus we find that $T_H \geq 26$, 2.6 and 0.26 ms, respectively, for sodium densities of 10^{11}, 10^{12} and 10^{13} atoms/cm^3.

For a useful target, the A–H spin exchange term $n_H\langle\sigma v\rangle(P_A - P_H)$ must be much larger than the alkali polarization loss term P_A/T_A so that the latter term can be ignored in eq. (8). We estimate how much larger the A–H spin exchange term is than the alkali polarization loss term as follows. The ratio of the A–H spin exchange terms to the alkali polarization loss terms is given by $n_H\langle\sigma v\rangle T_A(1 - P_H/P_A) = n_A\langle\sigma v\rangle T_H(n_H/n_A)(T_A/T_H)(1 - P_H/P_A) = (n_H/n_A)(P_H/P_A)(T_A/T_H)$. We are discussing the case of $P_H/P_A \geq 6/7$. A reasonable value of n_H/n_A might be 100 and T_A/T_H might be expected to be of the order of 1. Thus the ratio of the A–H spin exchange terms to the alkali polarization loss terms is expected to be about 80 so that almost all the angular momentum that is pumped into the alkali vapor is transferred into the hydrogen ensemble and only a small amount is lost by alkali relaxation or flow of alkali atoms out of the cell.

If in the steady state we consider $n_A V$ times eq. (8) and $n_H V$ times eq. (9), we find that

$$\frac{2Rn_{A\beta}V}{N} = n_A n_H\langle\sigma v\rangle V(P_A - P_H) + \frac{n_A P_A V}{T_A}$$

$$\simeq n_A n_H\langle\sigma v\rangle V(P_A - P_H) = \frac{n_H P_H V}{T_H}.$$

The rate at which angular momentum enters the alkali vapor by optical pumping is $2Rn_{A\beta}V/N$. The maximum rate at which angular momentum can enter the alkali vapor is the number of photons incident on the optical pumping cell per second divided by the average number of photons required to polarize an alkali atom in a high magnetic field, i.e. $R_{max}n_{A\beta}V/N = P_L/(h\nu N)$. In the expression P_L is the laser power. It follows that

$$\frac{2P_L}{h\nu N} \geq n_A n_H\langle\sigma v\rangle(P_A - P_H) \cong \frac{n_H P_H V}{T_H}. \quad (12)$$

Radiation trapping affects the rate at which polarized hydrogen atoms can flow out of the cell through the factor N^{-1} on the left-hand side of eq. (12). The effect of radiation trapping on spin exchange optical pumping in a high magnetic field occurs because radiation trapping increases the average number of photons that are needed to polarize an alkali atom. Radiation trappping occurs when the alkali vapor becomes optically thick so that multiple scattering of light is important for one or more radiative branches of the vapor. In a high magnetic field radiation trapping does not prevent the polarization of the alkali vapor, but it does increase the average number of photons needed to polarize an alkali atom [3]. This is in contrast to optical pumping in a low magnetic field where the multiple scattering of the photons results in a depolarizing mechanism for the vapor [4].

We now estimate the average number of photons needed to polarize a sodium atom for sodium densities of 10^{11}, 10^{12} and 10^{13} atoms/cm^3, when the alkali atoms are pumped using σ^+ light. The $^2P_{1/2}$, $m = 1/2 \to {}^2S_{1/2}$, $m = 1/2$ fluorescence is trapped if the population density of the $^2S_{1/2}$, $m = 1/2$ is sufficiently high. The optical absorption cross section between the $^2S_{1/2}$, $m = 1/2$ state and the $^2P_{1/2}$, $m = 1/2$ state in the alkali atom is given by

$$\sigma_{ABS} = \frac{\lambda^2}{8\pi}A_{ul}g(\nu - \nu_0), \quad (13)$$

where $\lambda = 599.6$ nm is the wavelength for the transition, A_{ul} is the Einstein A coefficient between the upper and lower levels, and $g(\nu - \nu_0)$ is the normalized Doppler line shape. At line center $\sigma_{ABS} = 4.4 \times 10^{-12}$ cm^2 at 500 K. We take the optical pumping cell to be a cylinder with a radius of $r = 1$ cm. Radiation trapping becomes important when $r \simeq (n_{A\beta}\sigma_{ABS})^{-1} = 2.3 \times 10^{11}/n_{A\beta}$, where $n_{A\beta}$ is in atoms/cm^3. Thus for sodium densities less than 2.3×10^{11} atoms/cm^3 radiation trapping is not significant. When radiation trapping is not significant, an average of 3 photons are required to polarize an atom. This occurs because after an absorption of σ^+ light on the average the $^2P_{1/2}$, $m = 1/2$ state decays to the $^2S_{1/2}$, $m = -1/2$ state two-thirds of the time and to the $^2S_{1/2}$, $m = 1/2$ state one-third of the time. As the sodium density increases above 2.3×10^{11} atoms/cm^3 the spontaneous emission from the $^2P_{1/2}$, $m = 1/2$ state to the $^2S_{1/2}$, $m = 1/2$ state becomes trapped and more than 3 photons are needed on the average to polarize a sodium atom. This means that on the average a longer time is required to absorb the photons to polarize the sodium atom. Tupa et al. [3] have presented calculations on the effects of radiation trapping on the optical pumping of an alkali vapor in a large magnetic field. Fig. 2 in Tupa et al. shows the polarization of a sodium vapor as a function of the time after the laser was turned on for several values of the sodium density. Since the time required to polarize the sodium vapor is directly proportional to the average number of photons required to polarize the sodium vapor, one can estimate the average number of photons required to polarize a sodium atom. The net result is $N = 3$, 5 and 60, respectively, for sodium densities 10^{11}, 10^{12} and 10^{13} atoms/cm^3.

Using the average number of photons needed to polarize a sodium atom it is possible to calculate $2P_L/(h\nu N)$ for a given pump laser power. In eq. (12) it was shown that $P_L/(h\nu N)$ is an upper limit on $n_H P_H V/T_H$. In order to make the discussion concrete, let us consider a pump laser power of 1 W. A 1 W laser at 589.6 nm emits 3×10^{18} photons/s. The values of $2P_L/(h\nu N)$ are, therefore, 2×10^{18}, 1.2×10^{18} and 1.0×10^{17} s^{-1}, respectively, for sodium densities of 10^{11}, 10^{12} and 10^{13} atoms/cm^3, respectively. For a hydrogen polarization of 0.75 the hydrogen loss rates can be calculated. Using eq. (12) one finds that $n_H V/T_H$ must be less than $2P_L/(h\nu NP_H) = 2.6 \times 10^{18}$, 1.6×10^{18} and 1.3×10^{17} s^{-1}, respectively, for sodium densities of 10^{11}, 10^{12} and 10^{13} atoms/cm^3.

In order that the flow of polarized hydrogen atoms be as large as is possible it is necessary to have T_H as short as possible consistent with maintaining a high hydrogen polarization, i.e. $T_H \geq 6/(n_A\langle\sigma v\rangle)$. For the minimum values of T_H and for a given volume V, one can calculate the maximum values for n_H. For an optical pumping cell with a volume of 40 cm^3 (a cylinder 1 cm in radius and about 13 cm long) the maximum values of the atomic hydrogen densities, n_H, are 1.6×10^{15}, 1.0×10^{14}, and 8.7×10^{11} atoms/cm^3, respectively, for alkali densities, n_A, of 10^{11}, 10^{12} and 10^{13} atoms/cm^3. If the hydrogen densities are higher than these values, then for a 1 W laser and for the minimum value of T_H, one cannot obtain an atomic hydrogen polarization of 0.75. It should here be noted that n_H should not be much smaller than the maximum value $n_H = P_l T_H/(h\nu NVP_H)$ since a smaller value means that the angular momentum pumped into the alkali cannot be transferred to the hydrogen at the maximum possible rate $P_L/(h\nu N) = n_A n_H\langle\sigma v\rangle V(P_A - P_H)$ unless n_A is increased above the value required by eq. (10).

Since $P_L/(h\nu N)$ is directly proportional to the laser pump power it is clear that the optimum value of the hydrogen density, which is given by $n_H \simeq 2P_L T_H/(h\nu NP_H V)$ increases linearly with P_L. This implies that the flow rate of polarized hydrogen out of the optical pumping cell can be increased linearly with the laser pump power provided the atomic hydrogen density can also be increased.

4. Conclusion on high field spin exchange optical pumping

In summary, there are several conditions that must be satisfied in order to polarize a hydrogen ensemble by spin exchange optical pumping. (1) If P_H/P_A is to be near one it is necessary that $n_A\langle\sigma v\rangle T_H \gg 1$. For a give alkali density n_A, this determines a minimum value of the hydrogen polarization loss time T_H. This condition assures that T_H is large enough that each hydrogen atom makes several spin exchange collisions with an alkali during the time T_H. (2) The hydrogen target is operated in an optimum manner when the angular momentum is transferred to the hydrogen ensemble as fast as it can possibly be pumped into the alkali vapor by optical pumping so that $P_L/(H\nu N) = n_A n_H\langle\sigma v\rangle(P_A - P_H)$. This requires that the angular momentum be lost by the hydrogen as fast as it can be pumped into the alkali vapor so that $P_H n_H V/T_H \simeq P_L/(h\nu N)$. As a result of these conditions, and for operation at the minimum value of T_H, one finds that $n_H \simeq P_L T_H/(h\nu NP_H V)$. It is desirable that the target operate with this minimum value of T_H in order that the flow rate of polarized hydrogen out of the cell could be maximized. The maximum possible rate at which angular momentum can be pumped into the alkali vapor is equal to the number of pump laser photons incident on the optical pumping cell divided by the average number of photons needed to polarize an alkali atom. In a high magnetic field the average number of photons needed to polarize an alkali atom is 3 at low alkali density and increases rapidly at alkali densities large enough that radiation trapping becomes important (i.e. for $n_A\sigma_{ABS}r > 1$). This implies that more hydrogen atoms per second can be polarized at an alkali density low enough that radiation trapping is not significant, rather than at a high alkali density. Of course, in order to operate with a low value of n_A, it is necessary that T_H be long enough that $n_A T_N\langle\sigma v\rangle \ll 1$ and that n_H be nearly equal to $P_L T_H/(h\nu NP_H V)$.

Finally it should be understood that for spin exchange optical pumping it is desired to produce the polarized hydrogen at the maximum rate. This is best accomplished in a high magnetic field at an alkali density low enough that radiation trapping is negligible so that one uses the smallest average number of photons to polarize each atom. On the other hand if one wishes to make a dense alkali target, such as the one currently used for the charge exchange target in the optically pumped polarized ion source, it is desirable to use high alkali densities so that radiation trapping cannot be avoided. Finally it should be noted that it may be useful to use an optically pumped spin exchange target for the charge exchange target in an optically pumped ion source.

5. High field optical pumping using linearly polarized light

Our calculations were carried out for σ^+ pumping radiation absorbed by the transition $^2S_{1/2}$, $m = -1/2 \rightarrow {}^2P_{1/2}$, $m = 1/2$, where on the average 3 photons are required to polarize an alkali atom in a high magnetic field. It may be better to use the π pumping radiation absorbed by the transition $^2S_{1/2}$, $m = -1/2 \rightarrow {}^2P_{1/2}$, $m = -1/2$, where on the average only 1.5 photons are required to polarize an alkali atom in a high magnetic field. In a high field, absorption of π light by the transition $^2S_{1/2}$, $m = -1/2 \rightarrow {}^2P_{1/2}$, $m = -1/2$ occurs at a different wavelength than for other transitions out of the ground level. The use of π radiation for the optical pumping means one must pump with a laser beam that is perpendicular to the magnetic field. It may also be useful to use the σ^+ absorption for the transition $^2S_{1/2}$, $m = -1/2 \rightarrow {}^2P_{3/2}$, $m = 1/2$, which also requires on the average only 1.5 photons to polarize an atom.

These are potential applications for optical pumping using π polarized light incident perpendicular to the magnetic field. One may be able to produce a long polarized target with a large value of nl parallel to the magnetic field. Also, the use of multiple lasers for the optical pumping of a target may be simplified. If one can produce a target with a large value of nl parallel to the magnetic field it may be possible to utilize collisional pumping for producing polarized ion beams [5].

6. Low field spin exchange optical pumping

Although the primary subject of this paper is spin exchange optical pumping in a high field, it may be of interest to consider briefly spin exchange optical pumping in a low field. In a low magnetic field, radiation trapping acts as a depolarizing mechanism so that one cannot polarize an alkali vapor with a density higher than $(\sigma_{ABS} r)^{-1}$, where σ_{ABS} is the level to level absorption cross section and r is the radius of the cylindrical optical pumping cell [4]. For a sodium vapor at 500 K, $\sigma_{ABS} = 4.4 \times 10^{-12}$ cm^2 at line center. For $r = 1$ cm the maximum sodium density that can be pumped in a low field is 2.3×10^{11} atoms/cm^3. At this density, in order that $P_A = 7/8$ and $P_H \geq 3/4$, one finds that $T_H \geq 2.6 \times 10^9 n_A^{-1} = 11$ ms. The average number of photons needed to polarize a sodium atom in a low field is 6. For a 1 W pump laser $P_L/(h\nu N) = 5 \times 10^{17}$ s^{-1}. This leads to

$$\frac{n_H V}{T_H} \leq \frac{P_L}{h\nu N P_H} = 6.7 \times 10^{17}\ \mathrm{s}^{-1}.$$

If T_H has the minimum value of 11 ms, then n_H must be less than 1.8×10^{14} atoms/cm^3.

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References

[1] K.P. Coulter, R.J. Holt, E.R. Kinney, R.S. Kowalczyk, D.H. Patterveld, L. Young, B. Zeidman, A. Zghiche and D.K. Toporhov, Phys. Rev. Lett. 68 (1992) 174.
[2] H.R. Cole and R.E. Olson, Phys. Rev. A31 (1985) 2137.
[3] D. Tupa, L.W. Anderson, D.L. Huber and J.E. Lawler, Phys. Rev. A33 (1986) 1045.
[4] D. Tupa and L.W. Anderson, Phys. Rev. A36 (1987) 2142.
[5] L.W. Anderson, S.N. Kaplan, R.V. Pyle, L. Ruby, A.S. Schlachter and J.W. Stearns, J. Phys. B17 (1984) L229.