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# RADIATION TRAPPING IN AN OPTICALLY PUMPED ALKALI VAPOR

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## ABSTRACT

Calculations are presented of the limitations imposed by radiation trapping on the electron spin polarization produced in an alkali vapor by optical pumping in a large magnetic field. It is found that electron spin polarizations of 90% are possible with Na densities up to  $10^{19}$  atoms/m<sup>3</sup> in a large magnetic field for ground level relaxation times of 150  $\mu$ s using a cylindrical geometry of radius  $7.5 \times 10^{-3}$  m.

The use of laser optical pumping to produce a polarized alkali vapor is of current interest. An important effect in the optical pumping of alkali vapors is the trapping of the pumping light [1]. Trapping occurs when the alkali vapor is sufficiently dense that multiple scattering of the light is important for one or more radiative decay branches of the vapor. This paper reports calculations of the effect of radiation trapping on the electron spin polarization produced in an alkali vapor by optical pumping.

Previous radiation trapping calculations have treated the case where the ground level is unpolarized [2,3,4,5,6]. In an optically pumped vapor, the populations of the individual states in the ground level are unequal. Radiation trapping calculations in such a vapor, therefore, must include the polarization and angular dependence of the emitted and reabsorbed resonance radiation.

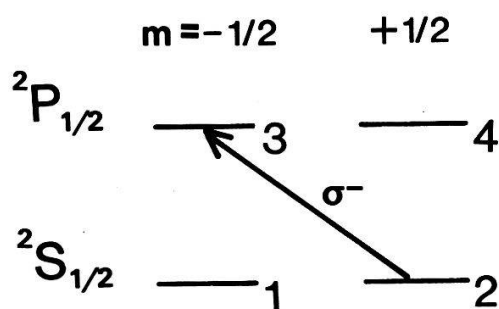


Fig. 1. Energy levels of the idealized alkali atom. Optical pumping with  $\sigma^-$  light excites the transition  $2 \rightarrow 3$  as shown.

We report calculations of the effects of radiation trapping for an "idealized alkali" atom with electron spin  $\vec{S}$  of  $1/2$  and with zero nuclear spin. We discuss the optical pumping of the idealized alkali atom using  $\sigma^-$  polarized light with a wavelength corresponding to absorption from the  $2S_{1/2}$   $m = 1/2$  state in the ground level to the  $2P_{1/2}$   $m = -1/2$  state in the lowest lying excited level.

Fig. 1 shows the relevant energy levels. For our calculations, we assume the optical pumping is carried out in a magnetic field large enough that the wave lengths  $\lambda_{13}$ ,  $\lambda_{14}$ ,  $\lambda_{23}$ , and  $\lambda_{24}$  are separated by several Doppler line widths so that the lines are completely isolated. We also assume that collisions of alkali atoms with other atoms or

with walls result in a relaxation between ground states 1 and 2 but do not result in appreciable excitation transfer between the excited states 3 and 4.

We calculate the populations of states 1, 2, and 3 as follows. The laser light at wavelength  $\lambda_{23}$  excites atoms from state 2 to state 3, and stimulates atoms in state 3 to emit and return to state 2. Atoms in state 3 may decay spontaneously to either states 1 or 2, and the spontaneously emitted photons can be trapped. If the population of atoms in states 1 and 2 are unequal, collisional relaxation occurs between these states. These processes are described by rate equations such as the equation for  $n_2$ , the density of atoms in level 2:

$$\begin{aligned} \frac{dn_2(\vec{r}, t)}{dt} = & - \frac{\lambda_{23}^2}{4\pi} A_3 \left[ \int \frac{I_{\vec{v}}}{h\nu} g(v_{23} - \bar{v}) d\bar{v} \right] [n_2(\vec{r}, t) - n_3(\vec{r}, t)] \\ & + A_{32} n_3(\vec{r}, t) - \frac{n_2(\vec{r}, t) - n_1(\vec{r}, t)}{T_1} \\ & - \left\{ \int d^3\vec{r}' d\nu n_3(\vec{r}', t) \frac{A_3(1+\cos^2\theta)}{8\pi|\vec{r}-\vec{r}'|^2} g(\nu - \nu_{23}) \right. \\ & \times \exp\left[-\frac{\lambda_{23}^2}{8\pi} A_3(1+\cos^2\theta) g(\nu-\nu_{23}) \int_0^{|\vec{r}-\vec{r}'|} (n_2-n_3) d\rho \right] \\ & \left. \times \frac{\lambda_{23}^2}{8\pi} A_3(1+\cos^2\theta) g(\nu-\nu_{23}) [n_2(\vec{r}, t) - n_3(\vec{r}, t)] \right\}, \quad (1) \end{aligned}$$

There are similar equations for  $n_1$ , and  $n_3$ . The Einstein A coefficients between state 3 and states 1 and 2 are, respectively,  $A_{31}$  and  $A_{32}$ . Thus  $A_3 = A_{31} + A_{32}$ , where  $A_3^{-1}$  is the radiative lifetime of state 3. For the decay

from a  $^2P_{1/2}$  level to a  $^2S_{1/2}$  level,  $A_{31} = A_3/3$  and  $A_{32} = 2A_3/3$ . The relaxation time between states 1 and 2 is  $T_1$ . The intensity per unit frequency of the light source used for the optical pumping is  $I_{\vec{v}}$ ;  $\bar{v}$  is the frequency of the pumping light. The optical absorption cross section from state 2 to state 3 for  $\sigma^-$  light of frequency  $\bar{v}$  is  $\lambda_{23}^2 g(v_{23} - \bar{v}) A_3 / 4\pi$ . The angle between  $\vec{r} - \vec{r}'$  and the direction of the magnetic field is  $\theta$ . We denote frequency by  $\nu$  so that  $\nu_{ij} = c/\lambda_{ij}$ .

The rate equations are very complex. We make a number of assumptions in their solution. First we assume that the intensity of the pumping light is constant rather than an exponentially decreasing function of position as the light propagates through the alkali vapor, and we assume that the light source is broad compared to the absorption line so that it completely covers the line with constant intensity per unit frequency,  $I_{\vec{v}}$ . We assume the alkali vapor is confined to a cylindrical volume of radius  $R$  having a length very large compared to  $R$ . Third we assume that scattered photons reaching  $r = R$  are completely absorbed, i.e., the target walls are non-reflective. Fourth and most importantly, we assume that the atomic densities  $n_1$ ,  $n_2$ , and  $n_3$  are independent of the position in the cylindrical volume.

Using the stated assumptions we have solved the simultaneous integral-differential equations as follows. The integrations over frequency are evaluated using Hermite integration [7] over the Doppler line shapes, the integration over  $r'$  is carried out exactly, and the integration over  $\theta$  is carried out using the Gaussian integration of moments [7]. The resulting differential equations for  $n_1$ ,  $n_2$ , and  $n_3$  are then solved as a function of time using numerical integration with the initial conditions  $n_1 = n_2 = N/2$  and  $n_3 = n_4 = 0$  at time  $t = 0$ , where  $N$  is the total density. We evaluate  $n_1$ ,  $n_2$ , and  $n_3$  using the following parameters. The temperature of the target is 600 K, the radius of the target is  $R = 7.5 \times 10^{-3}$  m, the mass of the alkali is  $M = 23u$ , the resonant wavelength is  $\lambda = 589$  nm and  $A_3 = 6.1 \times 10^7 \text{ sec}^{-1}$ . These are the parameters appropriate for the Na target used in the optically pumped  $H^-$  ion source.

We show selected calculations on several graphs. Figures 2 and 3 show the time dependence of the electron spin polarization of the ground level. The electron spin polarization is defined as  $P = (n_2 - n_1)/(n_2 + n_1)$ . The relaxation time  $T_1$ , the light intensity  $I$ , and the total density  $N$  are treated as parameters.

As expected for  $\sigma^-$  pumping light, the polarization as a function of the time decreases from its initial value of 0 toward its steady state value. The steady state value of the polarization is given by

$$P = \left\{ 1 + \frac{2}{T_1(A_{31} - R_{31})} \left[ 1 + \frac{A_3 - R_{32} - R_{31}}{\alpha} \right] \right\}^{-1} \quad (2)$$

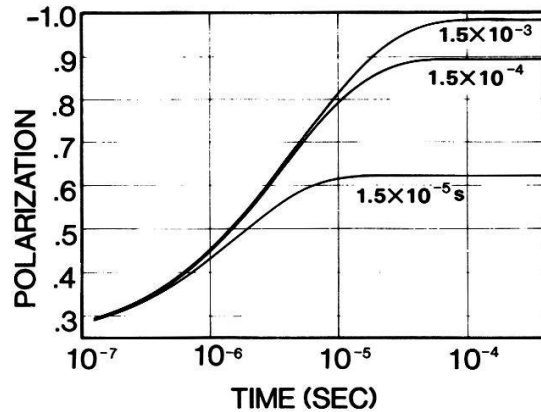
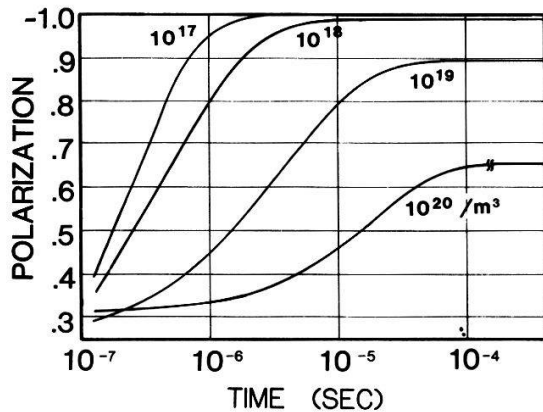


Fig. 2. Polarization as a function of time for different atomic densities  $N$ . The optical pumping begins with zero polarization at time  $t=0$ . Here we have taken the pumping intensity per unit frequency,  $I_{\nu}$ , to be  $2.85 \times 10^{-6} \text{ W/m}^2\text{-Hz}$  and the ground level relaxation time,  $T_1$ , to be  $150 \text{ } \mu\text{s}$ . A break in the scale for the atomic density of  $10^{20} \text{ atoms/m}^3$  is made to show the steady state polarization for that density.

Fig. 3. Polarization as a function of time for different ground level relaxation times  $T_1$ . The optical pumping begins with zero polarization at time  $t=0$ . These results are for pumping intensity per unit frequency,  $I_{\nu}$ , of  $2.85 \times 10^{-6} \text{ W/m}^2\text{-Hz}$  and atomic density,  $N$ , of  $10^{19} \text{ atoms/m}^3$ .

where  $\alpha = I_{\nu} \lambda_0^2 A_3 / 4\pi$ , and where  $R_{32}$  is the radiation trapping term in Eqn (1) divided by  $n_3$  and  $R_{31}$  is defined in a similar manner. In Table I we present the steady state polarization as a function of the intensity of the pumping light with the total density and the relaxation time treated as parameters. It is important to note that even when the optical pumping light intensity per unit frequency becomes infinite, the electron spin polarization of the optically pumped alkali vapor does not become large unless the spin relaxation time,  $T_1$ , is longer than the radiation trapped decay time from the excited state 3 to the ground state 1,  $(A_{31} - R_{31})^{-1}$ . In fact, for  $I = \infty$  we find  $P = -1[1 + 2/T_1(A_{31} - R_{31})]$ .

Table I. Steady state polarizations reached as a function of the ground state relaxation time,  $T_1$ , and the total atomic density,  $N$ . The cases shown are for pumping intensity per unit frequency,  $I_{\nu}$ , of  $7.13 \times 10^{-7} \text{ W/m}^2\text{Hz}$ ,  $2.85 \times 10^{-6} \text{ W/m}^2\text{Hz}$ ,  $1.14 \times 10^{-5} \text{ W/m}^2\text{Hz}$ , and  $\infty$ . These correspond to laser powers of 1/4 W, 1 W, 4 W, and  $\infty$ , respectively, evenly distributed over the cross sectional area of the cylinder and over the frequency bandwidth,  $g(0)^{-1}$ , which is nearly the Doppler linewidth.

		$I_{\nu}(\text{W}/\text{m}^2\text{Hz}) = 7.13\text{x}10^{-7}$	$2.85\text{x}10^{-6}$	$1.14\text{x}10^{-5}$	$\infty$
$T_1=1.5\text{ms}$	$N = 10^{17}/\text{m}^3$	.998	.9996	.9998	.99992
	$10^{18}$	.995	.998	.9993	.9996
	$10^{19}$	.958	.979	.989	.993
	$10^{20}$	.899	.913	.920	.929
$T_1=150\mu\text{s}$	$10^{17}$	.986	.996	.998	.9992
	$10^{18}$	.956	.985	.993	.996
	$10^{19}$	.841	.890	.918	.940
	$10^{20}$	.636	.657	.667	.681
$T_1=15\mu\text{s}$	$10^{17}$	.882	.961	.984	.992
	$10^{18}$	.770	.889	.941	.964
	$10^{19}$	.555	.622	.664	.708
	$10^{20}$	.281	.301	.312	.326

We have carried out various tests and believe that the approximations we have made do not affect the true solutions to these equations by more than about 20%. Ignoring nuclear spin causes us to underestimate the polarization that can be obtained at a given density so that our numbers probably represent a lower limit. We caution that our calculations should not be applied to the low field case.

There is interest in producing polarized alkali targets. For example a current use for an electron spin polarized alkali target is the optically pumped polarized  $\text{H}^-$  ion source [8,9,10]. One limitation on the density and polarization of the optically Na target is radiation trapping. For a sodium vapor target, relaxation times corresponding to 10-15 wall bounces have been obtained using a fluorocarbon rubber wall coating [11]. This corresponds, for our geometry, to 150  $\mu\text{s}$ . With this relaxation time, optically pumped Na vapor targets in a high magnetic field can have polarizations of greater than 90%, even for a vapor as dense as  $10^{19} \text{ atoms/m}^3$ .

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