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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^3 in a large magnetic field for ground level relaxation times of 150 μs using a cylindrical geometry of radius 7.5×10^{-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.

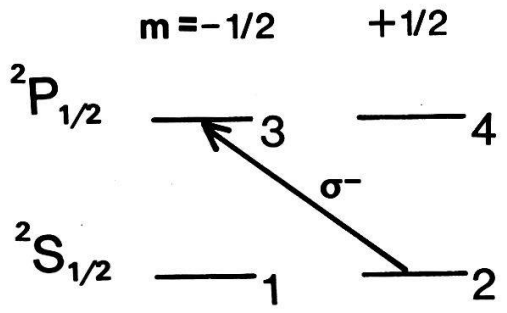

Fig. 1. Energy levels of the idealized alkali atom. Optical pumping with σ^- light excites the transition 2→3 as shown.

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 σ^- 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 λ_{13}, λ_{14}, λ_{23}, and λ_{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 λ_{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:

$$\frac{dn_2(\vec{r},t)}{dt} = -\frac{\lambda_{23}^2}{4\pi} A_3 \left[\int \frac{I_{\bar{\nu}}}{h\nu} g(\nu_{23} - \bar{\nu})d\bar{\nu}\right][n_2(\vec{r},t) - n_3(\vec{r},t)]$$

$$+ A_{32}n_3(\vec{r},t) - \frac{n_2(\vec{r},t)-n_1(\vec{r},t)}{T_1}$$

$$- \{\int d^3\vec{r}'d\nu\, n_3(\vec{r}',t) \frac{A_3(1+\cos^2\theta)}{8\pi|\vec{r}-\vec{r}'|^2} g(\nu - \nu_{23})$$

$$\times \exp[-\frac{\lambda_{23}^2}{8\pi} A_3(1+\cos^2\theta)\, g(\nu-\nu_{23}) \int_0^{|\vec{r}-\vec{r}'|} (n_2-n_3)d\rho\,]$$

$$\times \frac{\lambda_{23}^2}{8\pi} A_3(1+\cos^2\theta)\, g(\nu-\nu_{23})[n_2(\vec{r},t) - n_3(\vec{r},t)]\}, \qquad (1)$$

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 $I_{\bar{\nu}}$; $\bar{\nu}$ is the frequency of the pumping light. The optical absorption cross section from state 2 to state 3 for σ^- light of frequency $\bar{\nu}$ is $\lambda_{23}^2 g(\nu_{23}-\bar{\nu})A_3/4\pi$. The angle between $\vec{r}-\vec{r}'$ and the direction of the magnetic field is θ. We denote frequency by ν 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, $I_{\bar{\nu}}$. 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 θ 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 $R = 7.5\times10^{-3}$m, the mass of the alkali is M = 23u, the resonant wavelength is λ = 589nm and $A_3 = 6.1\times10^{7}\,sec^{-1}$. 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 σ^- 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

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

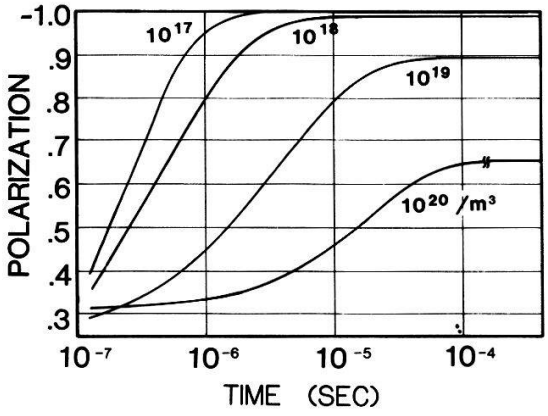

Fig. 2. Polarization as a function of time for different atomic densities N. The optical pumping begins with zero polarization at time t=0. Here we have taken the pumping intensity per unit frequency, $I_{\bar{\nu}}$, to be 2.85×10^{-6} W/m^{2}Hz and the ground level relaxation time, T_1, to be 150 μs.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

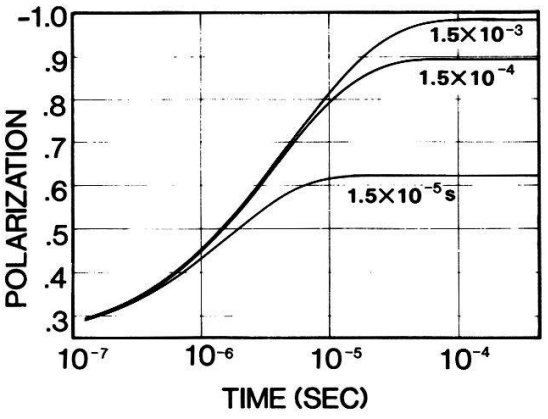

Fig. 3. Polarization as a function of time for different ground level relaxation times T_1. The optical pumping begins with zero polarization at time t=0. These results are for pumping intensity per unit frequency, $I_{\bar{\nu}}$, of 2.85×10^{-6} W/m^{2}Hz and atomic density, N, of 10^{19} atoms/m^3.

where $\alpha = I_{\bar{\nu}}\lambda_0^2 A_3/4\pi$, 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, $I_{\bar{\nu}}$, of 7.13×10^{-7} W/m^{2}Hz, 2.85×10^{-6} W/m^{2}Hz, 1.14×10^{-5} W/m^{2}Hz, and ∞. These correspond to laser powers of 1/4 W, 1 W, 4 W, and ∞, 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.

	$I_{\bar{\nu}}$(W/m^{2}Hz) =	7.13×10^{-7}	2.85×10^{-6}	1.14×10^{-5}	∞
T_1=1.5ms	N =10^{17}/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=150μs	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=15μs	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 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 μs. 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} atoms/m^3.

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